GEND-INF--019-Vol.2

DE83 011877

GEND-INF-019
Volume II

ESTIMATED SOURCE TERMS FOR RADIONUCLIDES AND SUSPENDED PARTICULATES DURING TMI-2 DEFUELING OPERATIONS PHASE II

Paul G. Voillequé
James R. Noyce
Charles A. Pelletier

Published May 1983

Nuclear Environmental Services
Science Applications, Inc.
Idaho Falls, Idaho
Rockville, Maryland

Prepared for EG&G Idaho, Inc.
and the U.S. Department of Energy
Three Mile Island Operations Office
Under DOE Contract No. DE-AC07-76ID01570

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 from TMI-2 and laboratory studies of relevant parameters were reviewed and used to develop estimates of source terms for total suspended particulates, particulate radionuclides, and soluble radionuclides during TMI-2 defueling operations. Potential control techniques for particulates and soluble species were reviewed and their design capabilities were estimated. Additional measurements that could reduce the uncertainty bounds for the estimates are recommended.

FOREWORD

This final report has benefited from comments provided by reviewers of the draft report and by attendees at review meetings. Written and oral comments were received from Bechtel National, Inc., Bechtel Northern Corporation, EG&G Idaho, Inc., GPU Nuclear Corporation, Oak Ridge National Laboratory, and Physical Sciences, Inc. The comments received aided our efforts to analyze a very complex problem. The authors remain responsible for the evaluations and interpretations given in the report and for any errors.

CONTENTS

TABLES

ESTIMATED SOURCE TERMS
FOR
RADIONUCLIDES AND SUSPENDED PARTICULATES
DURING TMI-2 DEFUELING OPERATIONS

INTRODUCTION

Normal refueling operations in a pressurized water reactor (PWR) involve flooding the refueling cavity around the reactor vessel with water. After the reactor head and plenum assembly have been removed and the cavity has been flooded, workers positioned on a bridge above the water are able to move fuel from one location to another in the reactor core and transfer spent fuel assemblies from the core to the cavity and then to the fuel storage pool. Concentrations of suspended solids and of radionuclides in the cavity water are normally quite low during refueling operations. Clear water in the cavity permits these manipulations and simultaneously shields the workers from the γ-radiation emitted by fission product isotopes contained in the fuel assemblies.

Removal of the fuel from the reactor vessel at Three Mile Island Unit Two (TMI-2) will be more complicated. Because the fuel was damaged during the accident, concentrations of radionuclides and suspended particulates in the cavity water will be significantly greater than for a normal refueling. The concentrations encountered can affect TMI-2 defueling operations in at least three ways. The potential problems are discussed in the context of normal refueling operations and procedures. First, the levels of suspended and dissolved radionuclides will affect radiation dose rates to personnel on the refueling bridge and airborne radionuclide concentrations. These in turn will dictate worker stay times and respiratory protection requirements. Second, suspended particulates may impair the workers' view of the fuel and cause delays until visibility is improved by natural processes or until turbidity control systems provide adequate removal. Third, 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, the cost of upgrading the refueling cavity cleanup system, and the total cost of defueling operations.

The primary goal of the study reported here was to develop engineering estimates of the source terms for soluble and suspended radionuclides and for visibility impairment due to suspended solids during TMI-2 defueling operations. These estimates are presented in the general context of a normal fuel handling arrangement, *i.e.*, access to the fuel from above with shielding provided by water in the reactor vessel and refueling cavity. Also considered are potential water turbidity and radionuclide concentration control techniques that could be employed during defueling. Thus the study provides information to those responsible for selecting defueling methods and specific subsystems needed to complete the work in an optimal manner.

In the next section, the original core and decayed inventories of the radionuclides most important to defueling operations are given. Indices that reflect the potential radiological hazards of these radionuclides are presented. The effects of the fuel damage, radioactive decay, leaching, and reactor coolant system (RCS) cleanup, are then considered to make estimates of the radionuclide distribution at the time of defueling. The next main section deals with the estimated amounts of suspended radioactive and stable particulates and potential means for turbidity control. This aspect is discussed first because the ability to see the fuel is an absolute prerequisite for wet defueling operations as described above, and because control techniques for particulates may increase concentrations of soluble radionuclides in the cavity water. Sources of and control techniques for soluble radionuclides are then considered. The last section contains recommendations for additional measurements that could help reduce the uncertainty bounds on the estimates that have been made.

INVENTORIES AND POTENTIAL HAZARDS OF RADIONUCLIDES

Radionuclide Inventories

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 comparable CINDER results are also in good agreement with those from the ORIGEN code.

In Table 1, the inventories are given for 18 isotopes of potential 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 small differences between the ORIGEN-2 and LOR-2 estimates for the other nuclides will not significantly affect the conclusions of this study. Most 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.

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

		Radionuclide Activity (Ci)[a]	
Radionuclide	Half-Life[b] (yr)	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	28.8	7.5×10^{5}	6.6×10^{5}
^{106}Ru	1.01	3.3×10^{6}	9.0×10^{4}
^{125}Sb	2.7	1.2×10^{5}	3.3×10^{4}
^{134}Cs[d]	2.062	1.6×10^{5}	2.7×10^{4}
^{137}Cs	30.17	8.4×10^{5}	7.5×10^{5}
^{144}Ce	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}
^{154}Eu	8.5	7.9×10^{3}	5.2×10^{3}
^{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 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.

d. Observed ratios of ^{134}Cs to ^{137}Cs in TMI-2 liquids were higher than would be expected from these shutdown inventories. If the inventory computed for ^{137}Cs is correct, then the calculated ^{134}Cs inventory is low by about 27%.

Indices of Potential Hazard

As indicated in the previous section, the nuclides shown in Table 1 have different characteristics and are important for different reasons. It is useful to rank the nuclides according to their potential to produce radiation exposure to personnel during defueling. Three categories of exposure were established (external gamma exposure, external beta exposure, and inhalation exposure), and the nuclides were ranked using an index of potential hazard for each category. The indices are simply indicators; they are not definitive measures of potential exposure. The three indices do not have a common basis and the numerical values for one category cannot be compared with those for another.

External Gamma Exposure

For the first category, external gamma exposure, the index was taken to be the product of the decayed inventory for a nuclide (Q_d, Ci) from the last column of Table 1 and its specific gamma-ray constant Γ ($m^2 \cdot R/hr \cdot Ci$). The specific gamma-ray constant for a radionuclide is numerically equal to the gamma exposure rate (R/hr) produced at a distance of 1 meter by a point source containing 1 curie of the radionuclide. The values of Γ, computed using data from Reference 3, and the product ΓQ_d are given in Table 2 for the gamma-emitting nuclides.

TABLE 2. INDEX FOR EXTERNAL GAMMA-RAY EXPOSURE

Radionuclide	Γ ($m^2 \cdot R/hr \cdot Ci$)	Gamma Exposure Index (ΓQ_d)
^{85}Kr	0.00127	8.8×10^1
^{106}Ru	0.108	9.7×10^3
^{125}Sb	0.243	8.0×10^3
^{134}Cs	0.874	2.4×10^4
^{137}Cs	0.318	2.4×10^5
^{144}Ce	0.0214	4.9×10^3
^{151}Sm	0.000498	5.5×10^0
^{154}Eu	0.562	2.9×10^3
^{155}Eu	0.0254	3.8×10^2

The values for the two radiocesium isotopes clearly dominate those for the other radionuclides. It should be emphasized that the indices are based on point source geometry and do not reflect self-attenuation by the fuel debris or other medium containing the radionuclides.

External Beta Exposure

The basic index for external beta exposure was taken to be the product of the decayed inventory (Ci) and average beta energy release per disintegration. $\bar{E}_\beta$ (MeV). The value of $\bar{E}_\beta$ for each nuclide was computed using data given in Reference 3 and is given together with the exposure index in Table 3. Also shown in Table 3 are bremsstrahlung exposure indices for the same radionuclides in fuel debris. The bremsstrahlung index reflects the fraction of the beta energy released that would be converted to photons, which are more penetrating than the beta particles. The bremsstrahlung index for a radionuclide in water would be about 9% of the index for fuel debris shown in Table 3. The dominant nuclides for either external beta or bremsstrahlung exposure are ^{90}Sr, ^{144}Ce, ^{106}Ru, and ^{137}Cs. These indices are based on the amounts of energy emitted during decay of the nuclides and do not reflect attenuation of the beta particles or photons in the potential source materials.

TABLE 3. INDICES FOR EXTERNAL EXPOSURE FROM BETA-EMITTING NUCLIDES

Radionuclide	$\bar{E}_\beta$ (MeV/dis)	Beta Exposure Index ($Q_d\bar{E}_\beta$)	Bremsstrahlung Exposure Index (Fuel Debris)
^{90}Sr	1.10	7.3×10^5	4.4×10^4
^{106}Ru	1.41	1.3×10^5	1.0×10^4
^{125}Sb	0.096	3.2×10^3	1.7×10^1
^{134}Cs	0.167	4.5×10^3	4.1×10^1
^{137}Cs	0.185	1.4×10^5	1.4×10^3
^{144}Ce	1.28	2.9×10^5	2.0×10^4
^{147}Pm	0.0699	5.7×10^4	2.2×10^2
^{151}Sm	0.0225	2.5×10^2	3.1×10^{-1}
^{154}Eu	0.240	1.3×10^3	1.7×10^1
^{155}Eu	0.0502	7.5×10^2	2.0×10^0

Inhalation Exposure

The index used to estimate the inhalation exposure potential for a radionuclide is the ratio of the decayed inventory activity (Q_d, Ci) to the annual limit on intake (ALI) (Ci) for that nuclide. Values of the ALIs for many radionuclides have been derived by the International Commission on Radiological Protection (ICRP).[4,5] The ALIs are given in Table 4 with values of the derived inhalation exposure index.

TABLE 4. INDEX FOR INHALATION EXPOSURE

Radionuclide	ALI (Ci)	Inhalation Exposure Index (Q_d/ALI)
^{3}H	8.1×10^{-2}	3.8×10^{4}
^{90}Sr	2.7×10^{-6}	2.4×10^{11}
^{106}Ru	1.1×10^{-5}	8.2×10^{9}
^{125}Sb	7.2×10^{-5}	4.6×10^{8}
^{134}Cs	1.1×10^{-4}	2.5×10^{8}
^{137}Cs	1.6×10^{-4}	4.7×10^{9}
^{144}Ce	1.4×10^{-5}	1.6×10^{10}
^{147}Pm	1.5×10^{-4}	5.6×10^{9}
^{151}Sm	1.8×10^{-4}	6.2×10^{7}
^{154}Eu	1.9×10^{-5}	2.7×10^{8}
^{155}Eu	8.1×10^{-5}	1.9×10^{8}
^{238}U	5.4×10^{-8}	5.0×10^{8}
^{238}Pu	5.4×10^{-9}	1.4×10^{11}
^{239}Pu	5.4×10^{-9}	1.7×10^{12}
^{240}Pu	5.4×10^{-9}	4.4×10^{11}
^{241}Pu	2.7×10^{-7}	6.7×10^{11}
^{241}Am	5.4×10^{-9}	3.5×10^{11}

The transuranic nuclides in the fuel debris have the highest values of the inhalation exposure index. The partially soluble fission products with high inhalation exposure potential are ^{90}Sr, ^{144}Ce, and ^{106}Ru.

Summary

The results of the hazard potential index calculations show that a short list of important radionuclides can be rationally compiled. The calculations indicate that the potential for external exposure of personnel is dominated by ^{137}Cs, ^{134}Cs, ^{106}Ru, ^{144}Ce, ^{90}Sr, and ^{125}Sb. Adequate control of the concentrations of these radionuclides in cavity water will minimize the potential external exposure of workers during TMI-2 defueling. It is worth remembering that the hazard indices for external exposure are based primarily on the radiation emission properties of the nuclei and that physical and chemical properties of the various elements and/or the material matrix will affect the actual risk to personnel.

The inhalation exposure potential is dominated by the transuranic radionuclides in the fuel debris. The short list of fission products given above is important as well because there may be a greater probability that they will be made airborne during wet defueling operations.

ESTIMATED RADIONUCLIDE DISTRIBUTION AT THE TIME OF DEFUELING

The expected distribution of radioactivity at the time of defueling depends upon (a) the available inventory, (b) core damage and radionculide release during the accident, (c) leaching of radioactivity from the fuel following the accident, and (d) leakage and processing of reactor coolant since the accident. The initial radionuclide inventory and that expected at the time of defueling were discussed in the previous section. The effects of the other processes are discussed in the subsections that follow.

Effects of Core Damage

General Effects

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.[6] 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 weakened but relatively 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 6. 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 32,000 to 58,000 kg. It has been estimated that core debris has blocked 60-90% of the area through which coolant normally flows upward in the core.[6]

The fuel damage that occurred was accompanied by substantial releases of volatile radionuclides. Large fractions of the noble gas, radioiodine, and radiocesium inventories 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 estimate the releases of fission products from the fuel. Initial release fractions for noble gases, tritium, radioiodines, and radiocesiums were estimated to be high.[7--9] The release fraction estimates are qualitatively consistent with the core damage estimates based on thermal-hydraulic and hydrogen generation calculations;[6] both approaches indicate substantial core damage.

The first video pictures from within the TMI-2 reactor vessel have generally confirmed that there was substantial damage to the fuel.[10] A bed of fuel debris was seen approximately 1.5 m below the center of the upper core plate. No definitive information on the size or other characteristics of the debris is presently available. No fuel rods were observed in a 0.7-m diameter circle where the center of the core had been. Insertion of the camera midway between the core axis and the outer boundary revealed that the ∿1.5-m deep void was also present at that location. The debris observed at that location was larger than was seen at the core axis. Insertion of the camera at the outer edge of the core was not possible, showing that some peripheral fuel rods were intact. It is not yet known whether the damage was symmetric around the core axis.

Effects on Cesium Inventory

Knowledge of the release of the cesium during the accident is particularly important to estimates of the potential radiation exposure during defueling. Using tank and reactor coolant concentration data and the results[11] of the August 1979 sump sample, Bishop et al. estimated that 60-70% of the radiocesium was released from the fuel, with the lower value, estimated using ^{137}Cs data, the more probable one.[7] Using the concentrations measured in small samples of the sump water that has been processed by the Submerged Demineralizer System (SDS) and data from EPICOR II[12] processing of water from the Auxiliary Building Tanks, Daniels et al. have estimated that only 36% of the radiocesium has been released from the fuel.[13] If the results[14] of the sump samples from Containment Entry #10 are used, the release fraction is estimated to be about 45%, using the procedure of Reference 13. The latter estimate is believed to be more valid. All these estimates of the radiocesium release fraction include both the initial release from the fuel during the accident and any leaching of radiocesium from the fuel over a period of several months thereafter.

An alternative approach is to calculate the release of cesium from the fuel during the period when the core was not adequately cooled. The expected release depends upon a number of parameters, but the most important is the temperature distribution within the core as a function of time.[15] Estimates of the temperature history of five sections of the TMI-2 core during the accident have been made using the Transient Reactor Analysis Code (TRAC).[16] Those temperature estimates were used together with release rate constants given in Figure 4.3 of Reference 15. These release rate coefficients were derived from experimental measurements of the total release from fuel at the end of a period during which the fuel was at a known temperature. Thus, the release rates are less well known than the total release and are considered to be order of magnitude estimates.[15]

The release of cesium from the fuel during the period between 10,000 s and 12,500 s (2.8 to 3.5 hours) after reactor shutdown was estimated for each of the five sections of the core considered in the TRAC calculation. The average temperatures during 500-s intervals, obtained from Figure 10 of Reference 16, were used to determine the release rate coefficient for each interval. The estimated cesium release fractions are shown in Table 5.

TABLE 5. ESTIMATES OF CESIUM RELEASE FROM THE FUEL, 2.8--3.5 HOURS AFTER SHUTDOWN

Level[a]	Fraction of Fuel in Each Level	Fraction of Cesium Released from Fuel	Fraction of Cs in Each Level
3	0.28	∼ 0	0.18
4	0.28	∼ 0	0.44
5	0.16	∼ 1	0.20
6	0.14	∼ 0.4	0.13
7	0.14	∼ 0.04	0.05

---

a. Corresponds to core divisions given in Reference 16. Level 3 contains the lowest portion (28%) of the core.

The fuel temperature estimates for the two lower levels given in Reference 16 imply that none of the cesium was released from the fuel located there. However, complete release of the cesium is estimated for the hottest region. It was assumed that the cesium distribution with height, in seven layers of nearly equal mass, could be adequately approximated by a cosine function that peaked in the center of the core and fell to zero at the top and bottom planes. The estimated cesium distribution is shown in the last column. Using columns 3 and 4, the cesium release during the period was estimated to be 25% of the inventory in the whole core. Most of this release from the fuel is estimated to have occurred in a period of about 10 minutes beginning 3.2 hours after shutdown. The high fuel temperatures were reduced dramatically by the high pressure injection flow about 3.3 hours after shutdown.

These calculations suggest that before core cooling was restored, approximately 28% of the cesium (including the gap inventory) had been released from the fuel. Approximately 70% of the fuel (Levels 3, 4 and 7) contained more than 95% of its initial inventory, about 16% of the fuel contained no cesium, and about 14% of the fuel contained 60% of its original inventory. Because the estimated fuel temperatures[16] during the period between 3.2 and 3.3 hours are so high, lowering the estimated temperature by 10% (230^{o}C) does not significantly change the release estimate for Level 5. Hence a lower bound release fraction is ∿0.2. Raising the peak core temperatures by 200^{o}C yields a release fraction of ∿0.4 after 3.3 hours.

The computed initial release fractions for cesium are lower than the fractional inventories of cesium found in the sump liquid. As noted above, this may be the result of leaching of cesium from the undepleted fuel. It may also reflect inaccuracies in the temperature estimates for the various core levels (or the appropriateness of the levels that were established in Reference 16) and corresponding release rate constants (Reference 15) that were used to compute the initial release. The fact that the axial and planar cesium distributions do not correspond to the fuel mass distributions may also affect the computations. Additional discussion of the difference between calculated and observed release fractions is given in the section on the effects of leakage from the reactor coolant system (RCS).

Effects on Strontium Inventory

The initial release of radiostrontium was estimated by Bishop *et al.*[7] to be <0.1%, using the initial RCS concentration. The low strontium release is one reason for the belief that fuel melting did not occur during the accident.[6] Subsequent measurements of both ^{89}Sr and ^{90}Sr in the RCS showed that the concentrations increased substantially and then remained relatively constant. Measured concentrations of ^{90}Sr in the reactor building sump[11,14] lead to estimates that 1.2--1.6% of the initial inventory had been found in liquids outside the core. Higher concentrations of ^{90}Sr were found in sump solids,[14] but the mass of solids is not known.

The initial release from strontium from the fuel during the period of inadequate core cooling was computed in the manner described previously, using fuel temperatures from Reference 16 and release rate constants from Reference 15. The estimated release from the fuel prior to core flooding at 3.3 hours after shutdown was 0.85% of the core inventory. As was the case for cesium, the estimate of strontium release from the fuel is lower than the amount of strontium estimated to be in various liquids. The range of estimates obtained by considering average temperatures that were 200°C lower and higher than the predicted values was 0.3 to 2% of the core inventory.

Effects on Inventories of Antimony and Ruthenium

Figure 4.3 of Reference 15 gives estimated release rate constants for two other elements whose isotopes are included in Table 1: antimony and ruthenium. The estimated releases from the fuel of these elements were computed using the predicted temperatures given in Reference 16. The predicted release fraction was 7.5% for antimony and 0.084% for ruthenium.

Isotopes of antimony and ruthenium could not be observed directly in liquid samples from the sump.[11] However, detectable concentrations of ^{125}Sb and ^{106}Ru were scavenged from the samples.[11] The observed concentrations correspond to 0.027% of the ^{125}Sb inventory and 0.00055% of the ^{106}Ru inventory. Both fractions are about two orders of magnitude lower than the predicted fuel release fractions. Both isotopes were detected in solids from the sump,[14] but the mass of solids is not known.

Effects of RCS Liquid Losses and Radionuclide Appearance Rates

The available data on radionuclide concentrations in reactor coolant can be used to estimate appearance rates for radiocesium and radiostrontium in the RCS and the effects of RCS discharges and leakage on inventories. Long term sequential RCS concentration data are not available for the other relatively insoluble particulate radionuclides listed in Table 1, so similar estimates for those nuclides are not possible. The appearance rates may reflect a number of processes. Radionuclides entering the RCS may have been leached from the fuel, dissolved from a precipitated compound, or removed from surfaces within the reactor vessel or primary piping. The calculated appearance rates are estimates of the total contributions of all processes.

The RCS has been sampled approximately once a week since the time of the accident. The sample obtained on 29 March 1982 was analyzed by Westinghouse Bettis Atomic Power Laboratory.[17] Routine analytical results for ^{3}H, ^{85}Kr, ^{89}Sr, ^{90}Sr, ^{134}Cs and ^{137}Cs in subsequent samples were reported by the Lynchburg Research Center of Babcock and Wilcox,[18] and, more recently, by GPU Nuclear.[19] Data on short-lived noble gases, radioiodines, ^{136}Cs and ^{140}Ba were reported when these radionuclides were present in significant quantities. Concentrations of chemical constituents of the RCS that have been determined routinely by the same laboratories are B, Na^+, Cl^-, H^+, O_2, H_2, and N_2. A few of the RCS samples have been split and analyzed by other laboratories to determine uranium and plutonium isotopic concentrations and the concentrations of fission product radionuclides.[20--22]

Most of the RCS samples are drawn from the letdown line in the cold leg of the RCS between the outlet of the A-loop steam generator 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, there is no forced coolant flow through the core and radionuclide decay has reduced fluid movement due to convection. The RCS makeup flow is injected on the outlet side of the A-loop reactor coolant pumps. Whether the limited convection, diffusion, and the leakage-makeup flows provide adequate mixing of the RCS is not certain. However, the data are suggestive: (a) results[18] from the few samples of the RCS taken from the pressurizer agree with those for the normal sampling location, (b) the rate of change of ^3H concentration in the RCS is generally consistent with the reported rate of RCS leakage, (c) the fact that the measured radiostrontium concentrations are relatively constant shows that ^{90}Sr activity entering the RCS has been transported to the sampling point, and (d) measured concentrations of cesium during reactor coolant processing operations were consistent with those calculated using the average liquid processing rates.[19] While none of the observations is unequivocal, the majority of the evidence supports the view that the coolant samples are representative of radionuclide concentrations in the RCS.

The radionuclide activities in the RCS were computed using the measured RCS concentrations that were decay corrected to the time of reactor shutdown. Equations (1) and (2) were used to describe the activity in the RCS during periods of relatively constant leakage. Equation (1) is a difference equation:

$$\frac{\Delta Q}{\Delta t} = P - \lambda_v Q \tag{1}$$

in which

ΔQ = the measured change in activity (Ci) in the RCS

Δt = the time period (days) when the change occurred

P = the average input to the RCS during the period, the appearance rate (Ci/day)

λ_v = the average fractional liquid loss rate constant (day^{-1}) for the period

Q = the average activity (Ci) in the RCS during the period.

Equation (2) is the integrated form of Equation (1):

$$Q = Q_0\, e^{-\lambda_v \Delta t} + \frac{P}{\lambda_v}\left(1 - e^{-\lambda_v \Delta t}\right) \tag{2}$$

where

Q_0 = the initial activity (Ci) of the radionuclide at the start of a time period.

Estimation of RCS losses and radionuclide appearance rates involves reconstruction of the fractional rates of liquid loss from the RCS during several time periods following the accident. The RCS leakage rates used for early time periods in this analysis are based upon a compilation of information on letdown and makeup rates from several sources,[23--26] and upon discharges from the pressurizer computed using the Moody critical flow tables and the RCS hotleg temperatures and pressures.[9] On 28 March, there were two periods of substantial discharge of coolant from the pressurizer. The first occurred during the first two hours and 18 minutes of the accident and involved relatively little transport of radioactivity out of the RCS because substantial fuel damage did not occur until about one hour later. From approximately three hours to 13 hours after the initiating event, there were periodic discharges for a total of 5.9 hours. Most of the later discharges occurred after the period of cladding failure, cladding oxidation, and cladding and fuel pellet fragmentation[16,27] when concentrations of radionuclides in the RCS were high. As noted in the previous section, the peak fuel temperatures are estimated to have occurred between 3.2 and 3.3 hours after shutdown.[16]

The RCS liquid loss rate constants used in the analysis are given in Table 6. The initial value of λ_v includes an average letdown rate of 60 gallons per minute (gpm) in addition to the average rate of discharge from the pressurizer during the period.[9] Discharges during the next four periods are based on plant data on RCS makeup. The majority of the water loss occurred during the first four months. The uncertainty of the individual values is estimated to be 10%. However, when the service water leakage and reactor building spray volumes are included, the sump water volume on 28 August predicted using Table 6 is within 5% of the 516,000 gallons present at that time.

The concentrations of ^3H in the RCS were used to estimate the liquid loss rate constants during the last two periods. The loss rate was obtained using a least squares fit for an exponential decrease during each period. The estimates in Table 6 assume that there were no significant additions of ^3H to the RCS during the two periods. The amount of ^3H remaining in the fuel at 120 days is estimated to have been <30% of the initial inventory. A total

TABLE 6. AVERAGE RCS LIQUID LOSS RATE CONSTANTS USED IN APPEARANCE RATE CALCULATIONS

Time Period	RCS Leakage Rate Constant λ_v (day^{-1})
3--13 hr[a]	5.9 ± .3
0.5--20 days	0.039 ± .004
21--35 days	0.036 ± .004
36--70 days	0.027 ± .003
71--119 days	0.0070 ± .0007
120--481 days	0.0030 ± .0003
482--810 days	0.0016 ± .0002
811--1145 days	0.0018 ± .0003

---

a. Discharges from the pressurizer prior to fuel damage did not contain significant quantities of ^{137}Cs or ^{90}Sr and were ignored in this analysis.

of 60% of the inventory was in liquids in the sump in tanks in the Auxiliary Building. An additional 10% was presumably released with the noble gases during the first three weeks after the accident and an undetermined amount was released in ventilation air after that time. The ^{3}H leaching data from the laboratory studies[28] are highly variable. The fraction of ^{3}H that would be leached during the period 120--810 days is estimated using data from Reference 28 to be 0.001 to 0.006 of the available inventory. If all the ^{3}H remaining in fuel is considered available, the the ^{3}H released from the fuel could be as high as ∿8 Ci in 690 days. The associated underestimate of λ_v between 120 and 480 days could be as high as 7%. In the last period, the value of λ_v could be 48% higher than the tabled value. Lower bounds on these errors can be estimated by assuming that the fraction leached during the period is 0.001 and that only half the ^{3}H in fuel is available. Under those conditions the errors due to the assumption that no ^{3}H was leached are within the estimated uncertainties for the values of λ_v. The loss rate for the last period is estimated by plant staff to be 0.1 gpm (0.0017 day^{-1}), which indicates that the estimate made using the ^{3}H concentration measurements is valid.

Table 7 contains the results of the calculations of the appearance rates of ^{137}Cs and ^{90}Sr in the RCS. The time periods used for the analyses for the two nuclides differ because the observed RCS concentration patterns differ. The appearance rates computed for the first time period for each nuclide are based on the assumption that the initial release from the fuel entered and mixed with the RCS ∿3 hours after shutdown.

TABLE 7. ESTIMATES OF APPEARANCE RATES FOR ^{137}Cs AND ^{90}Sr

^{137}Cs		^{90}Sr	
Time Period (days)	Appearance Rate P (Ci/day)	Time Period (days)	Appearance Rate P (Ci/day)
0.1--1.5	5.9×10^4	0.1--35	7.9×10^2
1.5--15	4.9×10^3	36--80	2.1×10^2
16--35	5.5×10^2	81--165	5.5×10^1
36-50	2.1×10^3	166--480	2.3×10^1
51--70	2.2×10^2	481--810	9.0×10^0
71--119	3.6×10^1	811--1145	3.8×10^0
120--480	1.1×10^1		
481--810	1.2×10^0		
811--1145	3.6×10^0		

It should be noted that the minimum detectable appearance rate is ∿2 Ci/day, so the estimated appearance rates for times greater than 480 days are quite uncertain. The uncertainties of the appearance rates for times before 480 days are estimated to be between 15 and 20% of the computed values.

The calculated appearance rates reflect releases from the fuel that are in addition to the initial release fractions, estimated above to be 28% for ^{137}Cs and 0.85% for ^{90}Sr. The additional release of ^{137}Cs is estimated to be 25% of the original inventory. For ^{90}Sr, the appearance rates in Table 7 imply an additional release of 7.1% of the original inventory.

It must be noted that the appearance rates reflect the time of entry of ^{137}Cs and ^{90}Sr into the RCS, not necessarily the time of departure from the fuel. That is, the entire release from fuel may have occurred between 2.8 and 3.5 hours, with deposition on surfaces and precipitate formation delaying the entry of the cesium and strontium into the RCS. This scenario is consistent with the dependence of release rate coefficients on temperature (Reference 15); however, it requires either the release rate coefficients (Reference 15), the computed temperatures (Reference 16), or the simplifying assumptions (or all three) used in the previous section to be incorrect.

Effects of RCS Processing

Processing of reactor coolant began 1146 days after reactor shutdown. Five batches of reactor coolant were processed between 17 May and 12 July 1982. The total volume of coolant removed and subsequently treated using the Submerged Demineralizer System (SDS) was ∿259,000 gallons. Flow rates out of the RCS ranged from 14.3 to 16.9 gpm. Samples of the RCS were collected at intervals ranging from ∿8 to ∿24 hours.[19] The effects of processing on ^{137}Cs and ^{90}Sr inventories in the RCS are discussed below. The effects of RCS processing after 12 July 1982 are not considered in this evaluation.

Effect on ^{137}Cs Concentration

The behavior of ^{137}Cs in the reactor coolant during processing was quite similar to that estimated using Equation (1) with $P \approx 0$ and λ_V computed using the weighted average letdown rate. The measured changes in concentration appeared to lag slightly behind those expected for perfect mixing. The ^{137}Cs activity removed from the RCS is estimated to be 4800 Ci, 0.6% of the original inventory. Approximately 0.07% of the inventory remained in the coolant after five processing campaigns.

There were increases in the ^{137}Cs concentration in the RCS between the initial processing campaigns. These were apparently due to activity from the pressurizer. The pressurizer was intentionally flushed during the fourth batch by directing the return flow of processed water through it. Following the fourth processing period, the calculated appearance rate was less than 1 Ci/day.

Effect on ^{90}Sr Concentration

The ^{90}Sr concentration in reactor coolant decreased by ∿50% during the first processing run but later returned to pre-processing levels. After the fifth campaign the concentration was again about half the original value. Approximately 11,000 Ci of ^{90}Sr, 1.5% of the original inventory, was removed during five processing runs.

The ^{90}Sr behavior supports the interpretation made early on by Malinauskas, Campbell, and others at Oak Ridge,[29] that the soluble ^{90}Sr is in equilibrium with a strontium compound that was precipitated in the primary system soon after release from the fuel. Filterable ^{90}Sr in the RCS samples was only ∿1% of the soluble activity at the end of the fifth processing run. The total ^{90}Sr activity available in the primary system to support the soluble ^{90}Sr level in the coolant is not known.

Effects of Radionuclide Leaching

Leaching of fission products from the fuel debris has occurred since the time of the accident. However, the amount of activity leached is not readily determined. Laboratory studies of radionuclide leaching from particles of irradiated fuel have been conducted under conditions that were similar in many respects to the TMI-2 situation. The results of these studies are discussed in this section.

The leach rates for a number of radionuclides have been measured experimentally under laboratory conditions, using samples of irradiated fuel.(28,30-32) The experimental leach rates from these studies are qualitatively similar. However, quantitative comparisons show the importance of particle size and leach solutions on the results. Solutions that have been used are deionized water, distilled water, Hanford groundwater, synthesized brine, and simulated pressurized water reactor coolant.

The study performed using simulated PWR coolant should be most relevant to leaching from the TMI-2 fuel. The solution contained ∿3300 ppm boron and ∿1100 ppm sodium at a pH of 8.[28] Measured values of the same parameters in the RCS at TMI-2 have averaged 3760 ppm boron, 1070 ppm sodium, and a pH of 7.8.[21,22] 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.[28] The oxygen content of the simulated PWR coolant was not controlled. This differs from TMI-2 where concentrations of oxygen in the RCS have been maintained at very low levels by chemical additions. Concentrations of elements other than boron and sodium were also not controlled and may have differed from the levels in the RCS at TMI-2.

Mitchell *et al.*[28] measured leaching rates and cumulative leaching fractions (amount leached/original inventory) for a period of 7.5 months. The relative rates of leaching found were Sb > I > ^{3}H > Cs > Sr > (Ru, Ce, Eu, and U) > Pu, a ranking that generally compared well with other studies. Ranges of the expected fraction of the available inventory that would be leached were extrapolated for periods up to four years. The estimates reflect differences in leaching for different particle sizes. Two principal fuel particle size ranges, between 14 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.[33] Some results are also given for other size ranges. Table 8 contains the extrapolated values of cumulative fractions leached after periods of 1 to 3 years for elements of interest for the TMI-2 defueling. Roughly 60 to 80% of the cumulative (3-year) leaching is estimated to occur during the first year. The measured leach rates dropped by an order of magnitude or more during the first three months of the experiments. The projected cumulative fractions leached are less than 1% for all elements except antimony. If the experiments performed using simulated PWR coolant are applicable to TMI-2, little of the radiocesium and radiostrontium released from fuel can be attributed to radionuclide leaching.

TABLE 8. ESTIMATES OF RADIONUCLIDE LEACHING FROM REFERENCE 28

	Estimated Range[a] for Percentage Leached During Period		
Element	1 Year	2 Years	3 Years
H	0.13--0.64	0.18--0.70	0.21--0.74
Sb	0.30--1.2	0.32--1.5	0.33--1.6
Cs	0.29--0.43	0.30--0.48	0.30--0.52
Sr	0.066--0.12	0.072--0.15	0.076--0.16
Ru	0.016--0.093	0.017--0.10	0.018--0.11
Ce	0.012--0.066	0.013--0.074	0.013--0.079
Eu	0.012--0.063	0.014--0.073	0.015--0.080
U	0.009--0.087	0.010--0.11	0.011--0.13
(Pu)[b]	0.004--0.022	0.005--0.026	0.005--0.030

---

a. The range reflects results for two different size fractions.

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

Estimated Radionuclide Distributions at Defueling

The preceding subsections lead to a number of conclusions that have an important bearing on the radionuclide distributions at the time of defueling. These conclusions are discussed below.

The initial releases of radionuclides from the fuel were estimated using calculated predicted fuel temperatures and release rate constants. The estimated values were 28, 0.85, 7.5, and 0.084 percent of the core inventories of cesium, strontium, antimony, and ruthenium, respectively. However, appearance rate calculations based on measurements of cesium and strontium in the RCS and information on RCS loss rates during the three years since the accident, showed that an additional 25% of the cesium inventory and 7.1% of the strontium inventory were released from the fuel into the RCS. In addition, laboratory studies of racionuclide leaching from fragmented fuel by simulated PWR coolant indicate that less than 0.6% of the cesium and less than 0.2% of the strontium releases would be due to leaching.

If it is assumed that the laboratory studies apply, then the initial releases from the fuel must have been ∿50% of the cesium and >8% of the strontium. These releases are double and ten times the corresponding estimates made using References 15 and 16. Approximately half of the initial cesium release was not initially seen in the RCS. This may have been due to deposition on surfaces during the period when the upper portion of the core and the plenum were filled with steam and hydrogen. Subsequent removal of this activity would then lead to the high appearance rates for ^{137}Cs in the RCS during the first few months following the accident. Most of the ^{90}Sr was also not immediately available. It is believed that a strontium precipitate formed and that its subsequent dissolution has supported the soluble ^{90}Sr concentration in the RCS.[29]

An initial strontium release of >8% requires higher fuel temperatures and greater core damage than estimated in References 16 and 27. Under those conditions, it would be expected[15] that a correspondingly larger fraction (∿20%) of the antimony would also have been released from the fuel. This has not been observed in the ^{125}Sb concentrations in liquids; however, the antimony could be bound to surfaces or present in solids.

The estimated ^{137}Cs appearance rates for the periods just prior to RCS processing, 481--810 and 811--1145 days after shutdown, are 1±2 and 4±2 Ci/day, respectively. Measurements made after the fourth RCS processing campaigns are consistent with an even lower cesium appearance rate, <1 Ci/day. Approximately 0.6% of the initial core inventory was removed by processing five batches of the RCS and about 0.07% of the initial inventory remained in the RCS.

The measured ^{90}Sr concentration changes during RCS processing support the view that the soluble ^{90}Sr in the RCS is supported by ^{90}Sr in solids in the primary system. The five processing campaigns removed 11,000 Ci of ^{90}Sr, ∿1.5% of the initial core inventory, while reducing the concentration in the RCS by about a factor of two. The ^{90}Sr activity that is available in solids is not known.

The results of the three insertions of the camera during the "quick look" suggest that the upper portions of approximately 80 of the fuel assemblies are no longer in place. It is possible that the tops of approximately 30 more assemblies were also severely damaged. However, it is also possible that those 30 assemblies are intact, as the outer two rows presumably are. (All these estimates assume symmetric damage around the core axis.) These estimates and the observed total cesium and strontium releases suggest that the core damage was between the reference and maximum damage estimates of Reference 6.

In estimating the radionuclide distribution at the time of defueling, the following mass fractions of fuel were used: loose fragmented debris, 0.4; fused debris, 0.1; relatively intact fuel pellets, 0.5. The estimated fractions of the original radionuclide inventory remaining in each category and the activity in that category are shown in Table 9. The estimated distribution of strontium, antimony, and cesium are rather uncertain. The estimated amounts remaining are consistent with the observed fractional releases from the fuel.

TABLE 9. ESTIMATED DISTRIBUTIONS OF IMPORTANT RADIONUCLIDES IN CORE DEBRIS CATEGORIES AT THE TIME OF DEFUELING

Radionuclide	Estimated (Fraction Remaining) and Activity (Ci) in Each Component: Loose Debris		Fused Debris		Fuel Pellets	
^{90}Sr	(0.7)	2×10^{5}	(0.9)	6×10^{4}	(1)	3×10^{5}
^{106}Ru	(1)	4×10^{4}	(1)	9×10^{3}	(1)	4×10^{4}
^{125}Sb	(0.6)	8×10^{3}	(0.8)	3×10^{3}	(1)	2×10^{4}
^{134}Cs	(0.1)	1×10^{3}	(0.3)	8×10^{2}	(0.9)	1×10^{4}
^{137}Cs	(0.1)	3×10^{4}	(0.3)	2×10^{4}	(0.9)	3×10^{5}
^{144}Ce	(1)	9×10^{4}	(1)	2×10^{4}	(1)	1×10^{5}
TRU-α [a]	(1)	6×10^{3}	(1)	1×10^{3}	(1)	7×10^{3}

---

[a] The sum of the activities of ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am, the alpha-emitting transuranic nuclides in Table 1.

The radionuclide concentration in the RCS at the time of defueling depends in part upon the number of additional RCS processing campaigns prior to defueling. Seven processing runs were scheduled originally, but it seems likely that additional time for processing will be available prior to head lift. It was assumed that three additional processing runs will be completed prior to defueling. Table 10 contains the projected inventory in the RCS and the gamma exposure index for each nuclide. As indicated, the inventories of ^{106}Ru, ^{125}Sb, and ^{144}Ce are not expected to exceed 1 Ci, but these projections are highly uncertain because little data on the concentrations of these isotopes in the RCS are available. Even after processing, the radiocesium isotopes dominate the potential gamma-ray exposure. The bremsstrahlung from ^{90}Sr is estimated to be the next most important source of gamma-ray exposure. The gamma-ray and bremsstrahlung contributions from the other nuclides are expected to be much less important.

TABLE 10. PROJECTED RCS INVENTORIES AND GAMMA EXPOSURE INDICES OF IMPORTANT RADIONUCLIDES AT THE TIME OF DEFUELING

Radionuclide	Q_{RC} (ci)	$\Gamma\ Q_{RC}$ (m^{2}R/hr)
^{90}Sr	$\sim 1\times10^3$	~ 5[a]
^{106}Ru	$\lesssim 1$	$\lesssim 0.1$
^{125}Sb	$\lesssim 1$	$\lesssim 0.2$
^{134}Cs	~ 5	~ 4
^{137}Cs	~ 70	~ 20
^{144}Ce	$\lesssim 1$	$\lesssim 0.002$

---

a. An approximation based on the expected fraction of emitted beta energy that would be converted to photons.

ESTIMATED SOURCES, CONCENTRATIONS, AND CONTROL OF SUSPENDED SOLIDS DURING DEFUELING

Background

Water clarity has been found to be a problem during routine refueling of light water reactors. 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. 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.[34]

The original designs of filtration and ion-exchange systems used for cavity water cleanup have been frequently found to be inadequate by power reactor plant operators. The inadequacies resulted from a lack of cleanup capability under certain conditions. Systems whose flow rates were too small for the volume of water treated ($\sim 4 \times 10^5$ gallons) were unable to reduce turbidity promptly. Solutions to the problems encountered have 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.[34]

The general operational experience cited above suggests that water clarity will be a serious problem during the defueling of TMI-2. In addition to the routine corrosion products responsible for visibility problems during normal operations, there are other major sources of particulates:

- Corrosion of metal surfaces in the primary system during and after the accident has produced an unknown mass of material that may enter the RCS during defueling operations.
- The bed of loose core debris contains fragmented fuel and cladding. The particle size of debris particulates has not been measured, but it is likely that a significant mass of particles will be <40 μm in diameter. Disruption of the debris bed, necessary for its removal,

will suspend some of the small particles present and may cause fragmentation that would increase the fraction of small particles.

- The bed of fused core debris will have to be separated into smaller pieces prior to removal from the reactor vessel. This process will entail cutting or fracturing the fused layer and will lead to releases of particulates.

Some specific examples of "nonroutine" problems with particulates in fuel pools have been documented.[35,36] These illustrate further the difficulties of reducing turbidity when unusual sources are present. Secondary problems associated with particulates include (a) deposition on cavity walls followed by increased beta- and gamma-radiation dose rates and/or resuspension of radionuclides into the air breathed by workers, (b) the formation of surface flocculants that increase dose rates and impair visibility, and (c) greater mechanical wear and degradation of components to be operated underwater.

The potential magnitude of the suspended particulate problem during TMI-2 defueling is addressed below. First, the data from analyses of RCS samples are presented and the changes observed during RCS processing are discussed. Then the possible generation rates during defueling and potential concentration reduction measures are considered. A point of reference for these discussions is a particulate corrosion product concentration of 1 ppm (1 mg/liter), a level that permits acceptable vision through ~30 feet of water[34] for normal PWR refueling. The concentration that can be tolerated depends on a number of specific parameters, including the type and size of the particles, the lighting, and the size of the object that must be manipulated. However, the value of 1 mg/liter can be used as a general guide for the concentration of particulates that could be tolerated without provision of equipment to aid operator vision.

Measurements of Particulate Matter in Reactor Coolant

The available information on particulate material in the reactor coolant system is mostly general in nature. Samples of the RCS have been described as "yellow" and "turbid" for about two years. However, the concentration of suspended particulates in the samples is not measured routinely. Gross turbidity measurements have been made since June 1982.

The concentrations and size distributions of suspended particulates have been reported for two samples of the RCS.[20] Part of the data reported for the samples called RCS-1 (collected 29 March 1979) and RCS-2 (collected 14 August 1980) are given in Table 11.

TABLE 11. DATA ON PARTICULATES IN THE RCS, MARCH 1979 AND AUGUST 1980, FROM REFERENCE 20

		Distribution of Particle Diameters (Fraction of Total Mass)		
Sample	Concentration (mg/liter)	>5 μm	1.2--5 μm	0.45--1.2 μm
RCS-1	68	0.49	0.21	0.30
RCS-2	128	0.0	0.15	0.85

The table shows two important features of the data that were markedly different in the two samples: (a) the total mass of suspended particulates in RCS-2 was nearly double that in RCS-1, and (b) the particles in RCS-2 are substantially smaller than in RCS-1. The median particle diameter was <3 μm in RCS-1 and <1 μm in RCS-2. It is not known whether particles that were originally large were fractured to produce smaller particles found in RCS-2 or whether the original particles settled and the small particles are the result of corrosion processes (or both).

The principal constituent identified in the solid portion of the 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 fraction with diameters between 0.45 and 1.2 μm) was analyzed, so the reported chemical constituents for that sample may be slightly biased. Concentrations of fissionable nuclides (U, Pu) in suspended solids also decreased substantially during the period between the two samples, suggesting that part of the > 5-μm fraction may have been dense fuel particles. The concentration of dissolved iron was much higher in RCS-2 than in RCS-1, suggesting increased corrosion and/or a greater injection rate of corrosion particulates.

There is insufficient data to determine whether sample RCS-2 represents a post-accident equilibrium particle distribution or whether conditions have changed since that time. It must be remembered that sample RCS-2 was collected nearly two years ago. The fact that the qualitative descriptions of the RCS samples have consistently been "yellow" or "turbid" may or may not reflect an underlying constancy of particle concentration and size distribution.

Effects of RCS Processing

Qualitative measurements of the turbidity of the RCS have also been obtained using a nephelometer, that is, by measuring the light scattered by the particles in the liquid sample. Unless the particle characteristics and a calibration curve have been established, this technique cannot be used to quantify the concentration of suspended solids. However the available data can be used to estimate the effect of RCS processing on turbidity, if it is assumed that the size distribution and character of the suspended solids were not changed in the process. The first five campaigns reduced the turbidity from ~ 36 nephelometric turbidity units (NTU) to 9.5 NTU. The fractional decrease in turbidity was less than that for radiocesium, suggesting that there was a continuous addition of particulate material during processing. The average particulate appearance rate during RCS processing can be estimated by making several assumptions. It was assumed that the nature and size distribution of particles was not changed, that the mass concentration at the start of processing was the same as in sample RCS-2, collected nearly two years earlier (see Table 11), and that changes in nephelometer reading are linearly proportional to concentration changes in this concentration range. Under the assumed conditions, the appearance rate for particulates during RCS processing was estimated to be ~ 1 kg/day.

The average appearance rate for the period between samples RCS-1 and RCS-2 was estimated using the leakage rate constants in Table 6 and the concentration data in Table 11. It is interesting, but perhaps fortuitous, that the estimated average appearance rate was ∿0.8 kg/day, consistent with the value estimated for the period of RCS processing. Corrosion rates measured under other conditions have been found to decrease with time,[37] so part of the addition of particles during RCS processing may have been due to disturbance of debris. The first video pictures from within the reactor vessel[10] showed that debris was easily disturbed. The movements of the camera in the guide tube frequently dislodged particulates.

It is expected that the planned additional RCS processing will reduce the suspended particulate concentrations even further. If the assumptions discussed above are valid and the appearance rate during subsequent processing is also ∿1 kg/day, then the concentration at the end of RCS processing is estimated to be about 16 mg/liter. If the appearance rate during quiescent periods between processing campaigns is greater than 0.01 kg/day then the final concentration could also be greater. The numerical estimates made in this section are of course highly dependent on the stated assumptions; however, they suggest that even after processing the particulate concentration in the RCS will substantially exceed the desired value of ∿1 mg/liter. Until other data are available, it is assumed that the particle size distribution will resemble that found in RCS-2. On that basis, the median diameter of suspended corrosion particles is expected to be less than 1 μm. The concentration may increase following processing as the result of debris disturbance prior to plenum removal.

Estimated Particle Additions During Plenum Removal

The amount of particulate material added to the RCS during plenum removal depends upon the difficulties encountered. As a minimum, part of the loose core debris that has settled onto plenum surfaces will be washed down into the RCS prior to and during plenum removal. Removal of these deposits will reduce both the β- and γ-dose rates and the potential for surface contamination and resuspension of radionuclides. As noted above, the video pictures[10] showed that there was a significant amount of loose debris on plenum surfaces. However, neither the mass of debris deposited on plenum surfaces nor the fraction that can be removed by water lancing is known. Settling velocities for the loose core debris (discussed in a later section) are substantial, so the addition of this material to the RCS is not expected to impair visibility for an extended period.

It may be necessary to cut the plenum into several pieces to remove it. Mechanical cutting or milling operations may be needed to relieve binding between the plenum and reactor vessel. The types and number of these operations will depend on the conditions actually encountered during plenum removal. Cutting may also be required to free the remaining end-fittings from the lower section of the plenum. The mass of cutting debris that will be generated is uncertain, but the particles are not expected to be small enough to impair visibility for an extended period.

Once the end-fittings have been freed, the lower plenum will not be attached to any core debris. Disturbance of the core debris, and a fraction of any cutting debris that has settled on top of it, should result only from the turbulence generated when the plenum is lifted. That will depend principally upon the rate at which the plenum is lifted out of the vessel. It is expected that, with a cautious lifting procedure, removal of the plenum will not greatly disturb the bed of loose debris that lies approximately 1.5 m (5 ft) below it. Any added particulate loading from lifting and water lancing during lifting is expected to be removed by settling within a short time after plenum removal.

Estimated Particle Generation During Removal of Core Debris

The total mass of loose and fused core debris is estimated to be between 32,000 and 53,000 kg.[6] Some of the fragmented core debris is small enough to have settled to the lower core support structure and bottom of the reactor vessel. The material observed in the plenum and in samples from the letdown filters strongly suggests that fine core debris will be found throughout the primary system. The initial attempt to probe the depth of the loose debris bed suggests that the bed is approximately 36 cm (14 in.) thick in the center of the reactor vessel. The top of the debris bed is 1.5 m (5 ft) below the core support plate at that location.[10] The moderator normally occupies 58% of the core[6] so the thickness of the bed could account for debris from about 86 cm (34 in.) of core height. A comparable amount of debris must be distributed (a) between intact fuel assemblies in the lower part of the core, or (b) in the bottom of the vessel, or (c) in the primary piping, pressurizer, and steam generators.

The fuel debris will be removed from the primary system during defueling. The preliminary defueling schedule suggests that defueling work will last about six months. The small particles of fragmented fuel and cladding will be encountered throughout the defueling period. While the removal of relatively intact stubs of fuel assemblies would not normally be expected to be a source of small particles, the small debris that has settled into the lower part of the core will be disturbed. Based on the preliminary schedule, the debris removal rate is estimated to be about 250 kg/day.

The effectiveness of the equipment used to remove the debris can be expected to be reasonably high, 80-99%. That is, the suction collection system would trap at least 80% of the material that is freed by application of mechanical force to the rubble, but it is unlikely to collect all of it. During some periods, the collection efficiency may be as low as 80% because forces applied in one area will disrupt material in a second location distant from the collection equipment. However, it is expected that an average collection efficiency in the higher portion of the range would be required. The average rate of release of debris bed material to the water would then be ∿1% of the debris removal rate. Hence, the nominal source term for core debris particulates is estimated to be ∿3 kg/day.

Potential Control Techniques for Particulates

It is clear from the previous discussions that turbidity control will be needed to improve water clarity during defueling. In this section, several potential control techniques are discussed. The effects of gravitational settling are considered first; then active removal processes are considered.

Gravitational Settling

Table 12 contains the terminal settling velocities for spherical UO_2 particles falling through water. The velocities were computed using Stoke's Law for water at $37^{o}C$ and a UO_2 particle density of 10.9 g/cm^3. For the less dense particles of ZrO_2 and iron oxides, terminal settling velocities under the same conditions would be smaller, about 46% and 42% of the tabled values, respectively. Terminal velocities for irregularly shaped debris particles would be greater than those shown for spherical particles.

TABLE 12. SETTLING VELOCITIES FOR SPHERICAL UO_2 PARTICLES IN WATER

Particle Diameter (μm)	Terminal Settling Velocity (cm/s)
1	7.8×10^{-4}
2	3.1×10^{-3}
4	1.2×10^{-2}
10	7.8×10^{-2}
20	3.1×10^{-1}
30	7.0×10^{-1}
40	1.2×10^{0}
60	2.8×10^{0}
80	5.0×10^{0}
100	7.8×10^{0}
200	3.1×10^{1}

---

a. Computed for spherical UO_2 particles with a density of 10.9 g/cm^3 falling through water at 37°C. Terminal velocities for spherical ZrO_2 and iron oxide particles would be about 46% and 42%, respectively, of the tabled values.

The time required for particle removal by gravitational settling can be quite large. Fine particles of UO_2 fuel debris, from 40 μm to 10 μm in diameter, will fall through 4.6 m (15 ft) of water in 7 to 45 minutes. However, the time required for 1-μm iron oxide corrosion particles to fall the same distance would be ∿19 days. These estimates ignore the effects of thermal currents, due to decay heat, that would retard settling. Gravitational settling cannot be relied upon to remove particles causing turbidity in the cavity water. The time scale is too great to permit efficient operations during defueling.

Directed Flow

During normal refueling outages, filling the cavity from above the core, rather than through the cold leg of the RCS, has been found to reduce the turbidity generated during cavity flooding. This is apparently because the mass of suspendable material attached to surfaces in the reactor vessel is relatively undisturbed when that procedure is used. Refueling experience has also 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.[34]

Similar flow patterns should be employed during TMI-2 defueling operations. Maintenance of a downward flow through the upper portion of the reactor vessel during defueling should minimize the dispersion of resuspended particles into the cavity water. Increasing the downward flow velocity at a physical interface between the cavity and reactor vessel by limiting the size of the opening would provide even greater confinement of the suspended particulates.

Supplementary Lighting and Viewing Equipment

Even though supplementary lighting and viewing equipment are possible measures to overcome, rather than control, vision problems caused by turbidity, it seems appropriate to discuss them briefly in this section.

Poor lighting for flooded pools and operational difficulties with installed systems have been common problems at operating plants.[34] Reference 34 suggests the use of long wavelength and polarized light to reduce scattering and the apparent turbidity. Directional, rather than general area, lighting is also recommended. A difficulty associated with installation of additional lighting is that it may encourage the growth of algae that can increase radiation levels by concentrating radionuclides in the cavity water and can also add to the turbidity problem.

It is almost certain that the normal refueling guide for particulate concentration (1 ppm) will be exceeded during substantial portions of the TMI-2 defueling. A moveable underwater viewing system--video camera with remote display screen--can be positioned near the work area to provide equipment operators with visual information required to perform their work.

Filtration

As indicated previously, skid-mounted and submersible filtration systems have been produced to supplement those supplied at the time of reactor construction. Such systems should be considered for application to turbidity problems expected during the defueling of TMI-2. 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 cleanup time considered acceptable. Determination of an acceptable cleanup time and optimum flow rate is beyond the scope of this work. The discussion that follows is only concerned with the capabilities of such systems, not their size or cost.

The equipment described in Reference 34 employs various types of filters: electromagnetic, etched disc, fabric bags, precoated cake on mesh, and the cartridge types. The cartridge, bag, and etched disc filters are available with ratings as low as 1 μm; typical operations have employed filters rated for particles approximately 5 μm in diameter. For submicron particles, such as those observed in sample RCS-2, it may be necessary to employ electromagnetic or cake filtration systems. The particle removal efficiency of an electromagnetic filter would depend upon the form of iron oxide present in the liquid under the conditions of operation.

Two conditions can limit the operation of filtration systems: <u>pressure drop</u> across the system due to the buildup of collected particulates and <u>radiation exposure</u> due to the radionuclides present in the particulate material. In normal use, the buildup of radionuclides has been more limiting than the filter mass loading, but that may not be the case for TMI-2. The time period required to change filters and the ease or difficulty in handling and disposing of the spent filter materials are important considerations in the selection of such systems.

The removal of particulates from the RCS or cavity water by filtration can be described using Equation (3):

$$\frac{dC}{dt} = \frac{P}{V} - \left(\frac{\varepsilon F}{V}\right) C \tag{3}$$

where

C = the particulate concentration (mg/liter)

t = time (s)

P = the rate at which particulates are entering the fluid (mg/s)

V = the fluid volume (liters)

ε = the removal efficiency of the filtration system

F = the flow rate through the filtration system (liters/s)

$\lambda_e = \varepsilon F/V$ = the effective removal rate constant for the filtration system (s^{-1}).

Equation (3) assumes that the dominant removal mechanism is the filtration system and that the system is well mixed. If either or both conditions were not met, a similar equation with a modified removal rate constant would apply. The general solution to Equation (3), when the initial concentration of particles is C_0 (mg/liter), is

$$C = C_0 \, e^{-\lambda_e t} + \frac{P}{V\lambda_e} (1 - e^{-\lambda_e t}) \tag{4}$$

The model described above can be used to estimate the effectiveness of filtration systems for removal of particulates under various conditions. However, it should be noted that a totally efficient filtration system (well mixed fluid and 100% collection of particles entering the filter) is not likely to be found. Reference 34 points out that actual cleanup half-lives have substantially exceeded design values under some operating conditions. No general rule appears applicable to describe reduced operational performance, but caution is necessary when discussing calculated system characteristics.

Under conditions when the appearance rate (P) is very small, the time (t^*, s) required to achieve a desired concentration (C^*, mg/liter) can be computed using Equation (5):

$$t^* = \frac{-\ln (C^*/Co)}{\varepsilon F/V} \quad (5)$$

In this case, the cleanup time is proportional to the liquid volume being treated, so confining the particulates to a small volume would shorten the time required to clarify the cavity water.

The more likely situation is that particulates will be entering the liquid more or less continuously during defueling. Under such conditions, the second term in Equation (4) controls the equilibrium particle concentration (C_e, mg/liter) given in Equation (6).

$$C_e = \frac{P}{\varepsilon F} \quad (6)$$

The equilibrium concentration of corrosion particles for P=11.6 mg/s (1 kg/day) entering a system with a continuous filtration rate of 0.95 liter/s (15 gpm) would be ∿12 mg/liter, if ε=1.0. The value of C_e for a continuous source of particles is independent of the liquid volume in which the particles are distributed. As shown earlier, settling would not significantly affect the concentration of these particles. Lower concentrations could be achieved by increasing the filtration rate. A completely efficient particulate filtration system with a flow rate of 11.6 liters/s (180 gpm) could maintain a corrosion particulate concentration of 1 mg/liter if the appearance rate of corrosion particles were 1 kg/day. As discussed above, supplementary lighting and viewing equipment are probably essential for the defueling operations.

The expected average injection rates for core debris are ∿3 kg/day. Removal of the larger and more dense core debris particles would be controlled by gravitational settling, not by cleanup system flow rate. However, provision of underwater video equipment is again desirable to avoid the delays that would be caused by disturbances of fuel debris during defueling.

Chemical Addition

Certain compounds of aluminum and iron are widely used as coagulants in water-treatment facilities. They react with the alkalinity of the water to form insoluble hydrated oxides (commonly called hydroxides), removing suspended particles in the process. The dosage varies, depending on the water conditions encountered, but is typically tens of mg/liter. High turbidity does not necessarily require a large dose of coagulant. The pH of the water must be kept sufficiently high by addition of an inorganic base during the process.

The choice of possible chemical treatments for reactor coolant and cavity water is narrowed by considerations of compatibility with materials in the reactor and the desirability of keeping the coolant non-acidic. The effectiveness of most of the possible treatments has not been demonstrated under TMI-2 conditions. Also, treatment to reduce turbidity might interfere with other water treatment processes. These caveats must be kept in mind when considering chemical modifications of the primary coolant and cavity water.

As indicated above, compounds containing chloride should be avoided (metal corrosion), and carbonate bases would evolve carbon dioxide if the reaction zone became acidic (possibly increasing airborne contamination). Two promising coagulants are dilute solutions of hydrated aluminum sulfate (alum) and anhydrous ferric sulfate. The second has the advantage of working effectively over a wider pH range: 5.5 to 8.3 versus 6.8 to 7.5 for alum.[38] However, alum is available as a concentrated solution that is convenient to use. The preferred bases to use to maintain alkalinity are solutions of calcium hydroxide (hydrated lime) or sodium hydroxide (caustic soda). It should be noted that moderate concentrations of sulfate or phosphate ion shifts the optimum pH toward acidity. Simplified chemical reactions for these two coagulants and these two bases are given in Table 13.

TABLE 13. SKELETAL EQUATIONS FOR REACTIONS OF METAL COAGULANTS WITH WATER ALKALINITY

$$Al_2(SO_4)_3 \cdot 14\ H_2O + 6\ NaOH \rightarrow 2\ Al(OH)_3 \downarrow + 3\ Na_2SO_4 + 14\ H_2O$$

$$Al_2(SO_4)_3 \cdot 14\ H_2O + 3\ Ca(OH)_2 \rightarrow 2\ Al(OH)_3 \downarrow + 3\ CaSO_4 + 14\ H_2O$$

$$Fe_2(SO_4)_3 + 6\ NaOH \rightarrow 2\ Fe(OH)_3 \downarrow + 3\ Na_2SO_4$$

$$Fe_2(SO_4)_3 + 3\ Ca(OH)_2 \rightarrow 2\ Fe(OH)_3 \downarrow + 3\ CaSO_4$$

To use these or other chemicals for effective water treatment they must be well mixed in the treated volume. As coagulation proceeds, flocs are formed which trap more suspended material as they settle. Mixing helps to form larger flocs, which remove particles more efficiently and settle in a reasonable time (up to a few hours). Flocculating aids will supplement, or diminish the need for, mixing. Of the several available, the two that seem most suitable are activated silica and polymeric flocculants, also known as polyelectrolytes. Activated silica is generally used with alum and can produce large, "tough" flocs that fragment less when disturbed after settling. The use of activated silica, however, is still somewhat of an art.[38] Polyelectrolytes are natural or synthetic organic polymers that promote the formation of very large, quick-settling flocs. Both kinds are available in anionic, cationic, or neutral types; the preferred kind and type for a particular situation must be determined empirically. The cationic type can be used alone but is not as effective (leaves more residual turbidity) as when used with alum. Together, they can flocculate algae and bacteria. Both activated silica and the polyelectrolytes are used in small quantities, typically 0.1 to 1.0 mg/liter. Excess amounts will actually disperse flocs. Bentonite clay in concentrations of 10 to 50 mg/liter is another flocculating aid that could be used.[38]

A system for out-of-vessel processing of refueling cavity water using these techniques was considered in Reference 34. In was generally considered too complex and messy for application to a normal refueling outage. Use of electromagnetic filters that employ the same flocculants to collect non-magnetic particles (or to improve the efficiency for hematite) was also not recommended for the same reasons.[34]

If the proper conditions can be achieved, coagulation and floccculation may be able to reduce coolant turbidity to the normal refueling guide value of 1 mg/liter. The flocs will coat opaquely the bottom surfaces they settle upon, thereby retarding the resuspension of loose particles that have previously sedimented. Such a process would probably have to be repeated several times during defueling. Not only will small particles be released by defueling activities, but also more particles might result from oxidation reactions when the vessel is opened. If this treatment process is pursued, careful selection and testing of candidate coagulants must be performed. The effectiveness of repeated applications should be evaluated experimentally prior to use. In addition, careful consideration of possible effects on other processes and on plant systems and materials is required prior to use.

ESTIMATED SOURCES OF RADIONUCLIDES AND THEIR CONTROL DURING DEFUELING

Two categories of sources are considered in this section, radionuclides in suspended particles and radionuclides in solution. The generation rates for particulates have already been discussed, so the extension to particulate radionuclides is considered first. Then sources of soluble radionuclides and potential control techniques are discussed.

Sources of Radioactive Particulates

Three principal sources of suspended particulates have been identified in the previous section: corrosion and core debris that have already been observed in the RCS, cutting debris generated during destructive plenum removal, and core debris disturbed during defueling. These sources are considered in subsections below.

RCS Suspended Particulates

The mass of small suspended particulates in the RCS at the time of defueling was estimated to be at least 5 kg. The actual value will depend upon the particulate appearance rate prior to and after the additional RCS processing campaigns. The total mass of suspendable corrosion particulates could be much larger. The fraction of the ^{137}Cs that was associated with particulates in sample RCS-2 was 0.24.[20] In more recent samples, the filterable ^{137}Cs fraction has been <1% of the total.[19] If that fraction remains constant, then <0.7 of the ~70 Ci of ^{137}Cs expected in the RCS at the time of defueling would be associated with particles, most of which would have diameters <1 μm. Similarly, <0.05 Ci of ^{134}Cs would also be in particulate form. The amount of ^{90}Sr in suspended particles is similarly estimated to be <10 Ci. Nearly all the ^{144}Ce in RCS-2 was in the particulate fraction. Neither ^{106}Ru nor ^{125}Sb was detected in RCS-2,[20] but the small amounts of these nuclides estimated to be in the RCS (Table 10) may be largely in the form of small particles.

Cutting Debris

As was noted previously, volatile radionuclides were released during the period when the plenum assembly was filled with gas and and were transported through the plenum prior to the discharge. Conditions were favorable for significant amounts of deposition on plenum surfaces:

- the surface to volume ratio of the plenum is large ($A = 9100\ ft^2$, $V = 1500\ ft^2$, so $A/V = 6.1\ ft^{-1}$ or $0.20\ cm^{-1}$)[39], and the deposition rate constant is proportional to A/V
- the flow rate of gas through the plenum (Q) was low, providing a long contact time (Q is estimated to have been $\lesssim 500\ ft^3/min$, so $Q/V \lesssim 0.33\ min^{-1}$ or $\lesssim 0.0056\ s^{-1}$)
- steam was condensing on plenum surfaces during the period.

These factors suggest that significant deposition of radioiodines and radiocesiums may have occurred during a period of $\sim$100 minutes following fuel cladding failure. Unfortunately, the deposition and resuspension rates and the rates of competing processes (e.g., scavenging by drops of condensate) are not sufficiently well known to predict the surface concentrations accumulated before the plenum was again filled with water. The deposited activity has been subject to leaching from the surfaces since that time.

To compute the potential contribution of radiocesium in cutting debris, the initial estimate of ^{137}Cs deposition on the lead screw surface was applied to plenum surfaces at the time of plenum removal. If that were the case, then the average plenum surface concentration would be roughly 1700 $\mu Ci/cm^2$. The ^{137}Cs activity released during possible cutting of plenum components is estimated to be less than 4 Ci. The particles generated by cutting are expected to be removed by gravitational settling, so the addition of $\sim$6% to the ^{137}Cs inventory would be temporary.

Core Debris

The average source term for loose core debris particles is estimated to be $\sim$3 kg/day throughout a 180-day defueling period. During short periods, the rate could be up to 50 kg/day. The estimated average rates of activity injection are shown in Table 14. The rates of injection for particles with diameters <40 μm assume that 15% of the debris lies in that size range. Both estimates employ average radionuclide concentrations based on the expected inventory in loose fuel debris (Table 9) distributed in 45 metric tons of fuel and cladding.

TABLE 14. AVERAGE RATES OF PARTICULATE RADIONUCLIDE INJECTION INTO THE RCS DUE TO DISTURBANCE OF LOOSE CORE DEBRIS

	Particulate Activity Injection Rate (Ci/day)	
Radionuclide	Loose Core Debris	Core Debris <40 μm
^{90}Sr	10	2
^{106}Ru	2	0.3
^{125}Sb	0.4	0.07
^{134}Cs	0.06	0.009
^{137}Cs	2	0.2
^{144}Ce	5	0.8

It is expected (Table 9) that the fused debris and intact fuel pellets will contain much higher concentrations of radiocesium and somewhat higher concentrations of ^{90}Sr than the loose core debris. However, the particulate injection rates for these debris categories are much lower. Unless the fused debris or intact rod stubs are pulverized, the mass of small particles produced during removal of these components should be small. For example, it is not expected that chiseling or cutting of fused core debris would inject small particles at a rate comparable to that for the loose core debris. Except for the previously discussed disturbance of loose debris, the removal of relatively intact stubs of fuel rods is likewise not expected to inject small particles into the reactor coolant.

Sources of Soluble Radionuclides

The most important source of soluble radionuclides during defueling operations is expected to be injection of radiocesium from fuel debris into the RCS. Injection of ^{90}Sr may continue depending, as discussed previously, upon the total amount available after RCS processing. However, if concentrations do not change substantially, Table 10 indicates that external radiation from ^{90}Sr will be a secondary concern. At existing levels, surface contamination by ^{90}Sr may be an operational problem. The potential for leaching of ^{106}Ru, ^{125}Sb, ^{144}Ce, and transuranics appears insignificant, but the potential for surface contamination and airborne concentrations of these nuclides cannot be ignored.

The radiocesium appearance rate during defueling may exceed that exhibited prior to RCS processing because the exposed surface area will be increased by disturbing the bed of loose debris, by cutting fused debris, and by fracturing fuel pellets. The vacuum collection system for loose debris may fragment the debris further and will wash the debris particles during collection. The radiocesium content of the intact pellets and fused debris is estimated to be several times higher than that in loose core debris (see Table 9); however, the potential for exposing additional surface area to water appears greatest for the loose debris. If the fused section and relatively intact section of the core can be separated by relatively simple cutting operations, little additional fuel area will be exposed. It is assumed that no significant pulverization of those debris fractions will occur.

The appearance rate of ^{137}Cs has decreased, following the fourth RCS processing campaign, to less than 1 Ci/day. This is lower than the estimated appearance rates just prior to processing (Table 7). On the basis of these measurements, the average ^{137}Cs appearance rate during defueling operations is estimated to be 0.5--2 Ci/day. An upper limit of 10 Ci/day is possible for brief periods.

The need for careful monitoring of the behavior of ^{137}Cs (and the other important radionuclides) during future RCS processing campaigns cannot be overemphasized. Better and more confident estimates of soluble radionuclide source terms during defueling will depend on additional good measurement data.

Potential Control Techniques for Radionuclides

A filtration system or other technique employed to control concentrations of suspended particulates would simultaneously reduce the amounts of particulate radionuclides. However, the soluble radiocesium source will probably be the most difficult to control, so additional treatment will be necessary. Potential control techniques are discussed below.

Demineralization of RCS and Cavity Water

Various types of demineralizer systems could be used for control of soluble radionuclides present in the RCS and refueling cavity water. As was the case for the filtration systems discussed previously, the system effectiveness depends upon the mixing and removal efficiency and the ratio of the flow rate to the volume of water being treated. Pressure drop across the system and limitations on the amounts of radioactivity collected are potentially controlling factors. The frequency of and time required for replacement of the ion-exchange medium may limit the operational effectiveness of such systems. The pressure drop and activity limits depend upon the system and resin transfer cask designs and are beyond the scope of this work.

The effectiveness of such systems can be assessed in the manner previously employed for filtration systems. Variants of Equations (3)--(6) apply, with the concentration and production terms given in mCi/liter and mCi/s, respectively. (It should again be noted that an epsilon of one may not be achieved in practice and that system operating efficiencies have been found to be lower than design values.[34])

Chemical Treatment of RCS and Cavity Water

Processes which remove the soluble species from solution so that they can be sedimented include precipitation, coprecipitation, occlusion, and adsorption. The measures previously described to control turbidity provide the additional benefit of reducing the concentration of some soluble radionuclides by coprecipitation. Those more effectively removed are cations with a valence state of 3, 4, or 5 (*e.g.*, ^{144}Ce). The efficiency is often greater than 50%, sometimes greater than 90%, when in slightly basic water (pH 7 to 8). Soluble Cs and Sr are only very slightly removed by these methods from water of low to moderate turbidity.

In water of higher natural turbidity, coagulation and flocculation removed some soluble Cs and Sr. Intentionally increasing turbidity by the addition of clay resulted in even more removal. The results from several laboratory studies are summarized in Table 15.[40] The results for ^{89}Sr should be applicable to ^{90}Sr. Added clay is not needed to precipitate trivalent ^{90}Y.

TABLE 15. LABORATORY DATA ON THE EFFECT OF CLAY ADDITION ON REMOVAL OF SOLUBLE CESIUM AND STRONTIUM

		Removal Fractions (%) for Clay Addition			
		Concentration of Clay Added (mg/liter)			
Radionuclide		0	100	750	5000
^{137}Cs	a.	0--6[a]	35--65 (38)[b]	-- (87)	-- (98)
	b.	0--37 (0.5)	--	--	--
^{89}Sr	a.	0--6	2--12	14--22	49--52
	b.	0--15 (3)	0--51	--	--

---

a. Range of observed values; not available for some tests.

b. Average values for a treatment are given in parentheses, not available for some tests.

The existing RCS turbidity could promote the removal of some of the soluble Cs and Sr as turbidity treatment proceeds. However, those suspended particles are not likely to have the adsorptive properties of clays. Any addition of clays should precede the introduction of coagulating agents. The recommended dose of clay as a flocculating aid, 10 to 50 mg/liter, is too low to be useful here. Note that hundreds of kg of sludge would result from additions of hundreds of mg of clay per liter of coolant.

Treatment with calcium oxide and sodium carbonate (lime and soda ash) at a concentration of 20 mg/liter can remove, under proper conditions, approximately 3/4 of soluble Sr. Higher concentrations removed more radiostrontium with about 99% sedimented following a 200-mg/liter treatment. This treatment is ineffective for the removal of Cs.[40] Possible carbon monoxide evolution is a negative aspect of this treatment.

Potassium dihydrogen phosphate (KH_2PO_4) and sodium phosphate have been reported to coagulate soluble ^{90}Sr. No data were presented for the effect on radiocesium.[40] Solid ammonium molybdophosphate will partly scavenge soluble ^{137}Cs at low (acidic) pH. It has little effect on radiostrontium. Apparently both treatments have been applied only on a laboratory scale. Neither treatment can be recommended because of the phosphate-caused pH shift toward acidity, as discussed earlier, and because phosphates promote the growth of algae.

Any proposed chemical treatment should be tested prior to application to assure that it will produce the desired results under TMI-2 conditions. In addition, careful consideration must be given to potential effects on plant systems and other processes.

Other Approaches

The difficulties associated with the two potential control techniques discussed above are substantial. Significant sedimentation of soluble radiocesium and radiostrontium without production of large amounts of sludge does not seem feasible. Use of demineralizers to maintain a low ^{137}Cs concentration would be expensive because of the large flow needed. To keep the dose rate on the refueling bridge (1.5 m above the water) at ∿50 mR/hour, the ^{137}Cs concentration in the cavity water must be maintained at ∿0.12 μCi/ml in the presence of an average source term of 1 Ci/day would be ∿1.6 gpm if the system effectiveness were 0.95. Partial isolation of the cavity water from the reactor vessel by maintaining downward flow through restricted openings in a physical barrier would reduce the cleanup requirements. In addition, local shielding may be devised to reduce dose rates in areas occupied by workers.

CONTROL OF THE GROWTH OF MICROORGANISMS

Some kinds of algae and bacteria can grow in darkness and without oxygen, but it is unlikely that any have survived in the coolant. Other algae grow best in bright light; the potential for their growth will increase when defueling operations begin. Phosphates are a major nutrient source, hence they should not be introduced into the RCS. Carbon dioxide promotes algal growth, thus coagulation treatments that release CO_2 should be avoided.

The use of chlorine or commercial chloride-containing aglicides and bactericides should probably be avoided because of possible corrosion effects. However, producers of proprietary compounds should be asked about the potential suitability of their other products. Bromine and iodine are bactericides, but their effectiveness in controlling algae is questionable.

Ozonation has been suggested[35] for use at TMI-2; however, ozone may be difficult to disperse in the RCS. The bubbling of an ozone-laden gas through the coolant seems undesirable because it probably would increase airborne contamination.

Copper, mercury, and silver metals are algicides at concentrations less than 1 ppm. Silver is also a bactericide and copper is slightly effective. Compounds of silver, such as $AgNO_3$, have also been used. However, turbidity-producing substances adsorb silver ion, rendering it ineffective. When cost is a consideration, cupric sulfate has been preferred; doses of less than 1 mg/liter are usually required. It should be added to water before any use of hydrated aluminum sulfate (alum) because alum efficiently removes copper.[41] All these compounds require contact times of hours to be effective. Potassium permanganate ($KMnO_4$) is reported to kill bacteria and algae at concentrations between 0.5 and 2 mg/liter. However, a 24-hour contact period is necessary for satisfactory results.[42]

If any chemical treatment process is pursued, careful selection and testing must be performed. The effectiveness of the treatment should be evaluated experimentally prior to use. Careful consideration of possible effects on other processes and plant systems is also required prior to use.

A treatment not requiring chemical additions to the water is irradiation with ultraviolet (UV) light. To minimize absorption losses, the irradiated water must be clear, so the UV irradiation cell should follow filtration in the processing sequence. The maximum bactericidal effect is achieved with UV light with wavelengths between 200 and 300 nm. Sterilization is normally accomplished by flowing a thin layer of water past the light source, but irradiation of water in a well mixed holding tank seems to be a reasonable alternative.

RECOMMENDATIONS FOR ADDITIONAL DATA COLLECTION

Collection of additional data would be useful for estimating the source terms during defueling. The following are recommended.

1. Carefully monitor the RCS concentrations of ^{90}Sr, ^{137}Cs, ^{144}Ce, and ^{106}Ru during future processing of the coolant by the Submerged Demineralizer System. Obtain additional samples of the RCS from the upper portion of the reactor vessel and perform detailed chemical and radiochemical analyses.

2. Obtain samples of loose core debris and measure the concentrations of radionuclides in them. Determine by gamma spectrometry the distribution of loose debris within the primary system, i.e., within the reactor vessel, the pressurizer, the steam generators, and in letdown and primary system piping.

3. Obtain by filtration particle size and suspended solids concentration information for additional samples of the RCS to determine whether the particle size distribution has stabilized or fluctuates. Determine whether the particles are abrasive and could damage equipment used for defueling.

4. Attempt to verify that the current low ^{137}Cs appearance rate will continue when the debris bed is disturbed. As a minimum, attempt a local disturbance with RCS samples collected nearby prior to, during, and after the disturbance.

5. Consider a test to verify mixing of the RCS. Injection of a tracer, such as ^{24}Na or ^{132}Cs, could provide the needed information without changing the chemistry of the RCS.

REFERENCES

1. R. G. Canada, NSAC EPRI ORIGEN Code Calculation of TMI-2 Fission Product Inventory, Technology for Energy Corporation Report. No. R-80-012, Knoxville, Tennessee (May 1980).

2. T. R. England and W. B. Wilson, TMI-2 Decay Power: LASL Fission-Product and Actinide Decay Power Calculations for the President's Commission on the Accident at Three Mile Island, DOE REPORT LA-8041-MS (October 1979).

3. C. M. Lederer and V. S. Shirley (eds.), Table of Isotopes, 7th Edition, New York: John Wiley & Sons, Inc. (1978).

4. International Commission on Radiological Protection, Limits for Intakes of Radionuclides by Workers, ICRP Publication 30, Parts 1 and 2 with Supplements, Oxford: Pergamon Press, 1979, 1980, 1981.

5. International Commission on Radiological Protection, Limits for Intakes of Radionuclides by Workers, Supplement A to Part 3 of ICRP Publication 30, Oxford: Pergamon Press, 1982.

6. D. W. Croucher, Three Mile Island Unit-2 Core Status Summary: A Basis for Tool Development for Reactor Disassembly and Defueling, DOE Report GEND-007 (May 1981).

7. W. N. Bishop, D. A. Nitti, N. P. Jacob, and J. A. Daniel, Fission Product Release from the Fuel Following the TMI-2 Accident, in Proceedings of the ANS/ENS Topical Meeting on Thermal Reactor Safety, Conference Report CONF-800403 (June 1980).

8. C. A. Pelletier, C. D. Thomas, Jr., R. L. Ritzman, F. Tooper, Iodine-131 Behavior During the TMI-2 Accident, EPRI Nuclear Safety Analysis Center Report NSAC-30 (November 1981).

9. C. A. Pelletier, P. G. Voillequé, C. D. Thomas, Jr., J. A. Daniel, E. A. Schlomer, and J. R. Noyce, Preliminary Radioiodine Source Term and Inventory Assessment for TMI-2, SAI-139-82-04-RV, Science Applications, Inc., Rockville, Maryland (April 1982).

10. GPU Nuclear, Recorded Video Pictures Taken Within the TMI-2 Reactor Vessel and Related Documentation, Unpublished (July 1982).

11. W. D. Schults, Letter report of Analytical Results for Sump Sample, Analytical Chemistry Division, Oak Ridge National Laboratory (September 1979).

12. U.S. Nuclear Regulatory Commission, Environmental Assessment, Use of EPICOR-II at Three Mile Island, Unit 2, NRC Report NUREG-0591 (August 1979).

13. R. S. Daniels, D. D. Miller, and J. M. Rodabaugh, Refueling Canal Cleanup System for TMI-2 Core Recovery, Bechtel National Inc., Draft Report (November 1981).

14. P. D. Randolph, First Results of TMI-2 Sump Sample Analyses--Entry #10, Letter (Rand-10-81) Report of Analytical Results, EG&G Idaho, Inc. (July 1981).

15. U.S. Nuclear Regulatory Commission, Technical Bases for Estimating Fission Product Behavior During LWR Accidents, NRC Report NUREG-0772, Nuclear Regulatory Commission, Washington, D.C. (June 1981).

16. J. R. Ireland, T. R. Wehner, and W. L. Kirchner, Thermal-Hydraulic and Core-Damage Analyses of the TMI-2 Accident, Nuclear Safety, 22, 583 (1981).

17. O. J. Woodruff, Three Mile Island Unit 2 Assistance, Letter report of Analytical Results, Westinghouse Electric Corporation (May 1979).

18. Babcock and Wilcox, Reactor Coolant System Sample Results, Periodic letter reports of Analytical Results (1979--1981).

19. GPU Nuclear, Data Sheets with Analytical Results (1981--1982).

20. C. W. Filby, TMI RCS and RCBT Sample Report, Letter (CWF-14-81), Report of Analytical Results, Exxon Nuclear Idaho Company, Inc. (June 1981).

21. J. E. Cline, G. J. Duggan, J. R. Noyce, K. L. Wright, and R. Lubaszewski, Characterization of TMI Unit 2 Post-Accident Primary Coolant, Science Applications, Inc. (August 1981).

22. R. L. Nitschke, TMI RCS-RCBT Report, Letter (RLN-2-82), Report of Analytical Results, EG&G Idaho, Inc. (January 1982).

23. S. Lamana, Water Inventory as of 0800, 3/30/79, Memo Report, Metropolitan Edison Company (May 1979).

24. Metropolitan Edison Company, Three Mile Island Unit 2 Recovery, Quarterly Progress Report for the Period Ending March 31, 1980, Annotated Sequence of Events, TMI-II-RR-6 (April 1980).

25. U.S. Nuclear Regulatory Commission, Investigation Into the March 28, 1979 Three Mile Island Accident by Office of Inspection and Enforcement, NRC Report NUREG-0600 (August 1979).

26. Nuclear Safety Analysis Center, Analysis of Three Mile Island - Unit 2 Accident, EPRI Nuclear Safety Analysis Center Report NSAC-80-1, NSAC-1 Revised (March 1980).

27. M. Rogovin et al., Three Mile Island--A Report to the Commissioners and to the Public, NRC Report NUREG/CR-1250 (January 1980).

28. A. D. Mitchell, J. H. Goode, and V.C.A. Vaughen, Leaching of Irradiated Light-Water Reactor Fuel in a Simulated Post-Accident Environment, DOE Report ORNL/TM-7546 (May 1981).

29. A. P. Malinauskas and D. O. Campbell, Oak Ridge National Laboratory, Personal communications (1981).

30. Y. B. Katayama, D. J. Bradley, and C. O. Harvey, Status Report on LWR Spent Fuel IAEA Leach Tests, DOE Report PNL-3173 (March 1980).

31. Y. B. Katayama and D. J. Bradley, Long Term Leaching of Irradiated Spent Fuel, DOE Report PNL-SA-7734 (November 1979).

32. Y. B. Katayama, *Spent LWR Fuel Leach Tests*, DOE Report PNL-2982 (April 1979).

33. C. D. Hodgman (ed.), *Handbook of Chemistry and Physics*, 40th Edition, Cleveland: Chemical Rubber Publishing Company, 1959.

34. D. P. Bour, D. S. Langston, W. Lechnick, and J. M. Riddle, *Refueling Outage Water Clarity Improvement Study*, EPRI Report NP-1081 (May 1979).

35. D. W. Rhodes, *ICPP Fuel Storage Pool Experience as Applied to TMI-2 Pool Cleanup*, Letter Report (Rhod-7-81), Exxon Nuclear Idaho Company, Inc. (June 1981).

36. C. R. Gerstberger, Jr., *Plant Information*, Letter (FS-CRG-430) Report on Operational Experience at Connecticut Yankee, Westinghouse Electric Corporation (July 1981).

37. W. E. Berry and R. B. Diegle, *Survey of Corrosion Product Generation, Transport, and Deposition in Light Water Nuclear Reactors*, EPRI Report NP-522 (March 1979.

38. J. M. Cohen and S. A. Hannah, Coagulation and Flocculation, Chapter 3 in *Water Quality and Treatment*, Third Edition, New York: McGraw-Hill (1971).

39. G. Thomas, Electric Power Research Institute, Personal Communication with C. A. Pelletier (November 1981).

40. C. P. Straub, Radioactivity, Chapter 13 in *Water Quality and Treatment*, Third Edition, New York: McGraw-Hill (1971).

41. C. V. James, The Removal of Colour, Odour and Taste, Chapter 3 in *Water Treatment*, Third Edition, Cleveland: CRC Press (1971).

42. E. J. Laubusch, Chlorination and Other Disinfection Processes, Chapter 5 in *Water Quality and Treatment*, Third Edition, New York: McGraw-Hill (1971).