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THE IMPACT OF DECONTAMINATION ON LWR
RADIOACTIVE WASTE TREATMENT SYSTEMS*

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INTRODUCTION

Increased radiation levels around certain reactors in the United States and accompanying increases in personnel exposure are causing a reexamination of available options for continued operation and performance of required maintenance and inspections. One of the options is decontamination of all or part of the primary system. Any decision to proceed with decontamination could have profound effects on the radwaste treatment system. This led the Nuclear Regulatory Commission (NRC) to sponsor work at the Pacific Northwest Laboratory (PNL), Battelle Memorial Institute, to identify and assess these effects.

A prerequisite to this discussion is a well understood definition of what is meant by decontamination. Decontamination as considered in this report encompasses all operations that have to do with the chemical or mechanical cleaning of a primary system or components of that system, and any unusual manipulation of the primary system water chemistry that would result in the planned and beneficial transport of radioactive materials within the system. This definition is more inclusive than those that are normally used to define decontamination. A broader definition, however, is needed because the focus of this study is on the impact of any processes which remove radioactivity on radwaste treatment systems.

An assessment of the impact of decontamination on radwaste treatment systems involves a careful study of the various effects

that would result from the use of such a process. Criteria are discussed that establish a context for what the authors believe could occur as a result of a decontamination.

The criteria used are:

- One measure of the impact on the radwaste treatment system is an assessment of the corrosion of radwaste system materials.
- What is the effect of decontamination on the amount of waste that must be processed and disposed of?
- Are the various unit operations laid out in such a way that the operating staff can accommodate higher radiation levels?
- Is there sufficient volumetric storage capacity?
- Will the processing of concentrated chemical solutions result in deposits on heat transfer surfaces that will impede operation or cause equipment failures?
- What are the effects on personnel exposure?
- Are processes currently available for solidifying decontamination wastes satisfactory?
- High concentrations of radioactivity have an impact on methods of transportation and disposal of the radwaste. Is onsite storage a realistic option?
- Safety analyses must cope with possible chemical burns, degradation of equipment, airborne material, hydrogen gas generation, etc. What features are pertinent to the system being examined?
- Costs of system modification or new systems must be considered. Why is decontamination being considered?
- Will additional personnel be needed?
- Is the technology now available to the utility for use in the radwaste treatment system adequate to accommodate decontamination?

Before addressing the specific impacts of each type of decontamination process, a discussion of the basic assumptions used in our radiation exposure calculations is needed.

In order to avoid confusing the issue, no man-rem estimates will be made when discussing radiation exposure resulting from handling radwaste from the various decontamination processes. An

estimate of the occupational doses associated with processing the decontamination waste, man-hour estimates can be made. These estimates, however, might not reflect reality and would, therefore, make the meaning of the exposure problems unclear. Hence, the radiation exposure rates for the various decontamination options are calculated in roentgens per hour (R/h). Where possible, these exposure rates are compared to rates which are reasonable during normal radwaste system operations. This procedure may better characterize occupational dose rates and identify problem areas which may be significant in terms of occupational exposures.

The radionuclides which are expected to contribute almost the entire radiation field during most decontamination waste processing are ^{58}Co and ^{60}Co ¹⁻⁵. Most of the radiation exposure rate calculations done for the decontamination waste are based on radiations emitted from ^{60}Co .

THE IMPACT OF DECONTAMINATION

The following discussion will consider the impact on the radwaste treatment system of each type of decontamination process, i.e., mechanical, dilute and concentrated. The criteria outlined above will be discussed as they apply to each of these processes.

THE IMPACT OF MECHANICAL DECONTAMINATION

Mechanical decontamination processes are characterized by use of fluids, fluids with abrasives or abrasive techniques. For any sizable operation, one or both of the first two techniques would be employed.

Mechanical decontamination is generally performed in water or with water for two reasons. The first is to minimize the amount of airborne contamination. The second is that water is a good carrier for abrasives and a ready means for transferring kinetic energy into a film removal action. The impact, therefore, falls mainly on the liquid and the solid radwaste systems with no significant impact on the gas system. The composition of the waste from the decontamination will be particulate corrosion products, probably with metal particles from the underlying structure, and if an abrasive is used, abrasive particles.

The first unit operation in the liquid system is a settling tank to remove the large particles. These larger particles are highly abrasive and could damage piping and components. After passing the pump, the waste would be filtered to remove suspended particles. The filtrate would enter the standard liquid radwaste system; it is not expected to have any special properties that would affect the system.

At the present time, there is not enough information to do a quantitative analysis of the radiation exposures associated with mechanical decontamination. Because this method of decontamination may be used on an isolated area of the reactor primary system, the waste stream will probably be less in volume than that for a concentrated decontamination. The mechanical decontamination process, however, will likely be used on areas which have high levels of radioactive crud buildup. This will likely result in a very high concentration of radioactivity in the waste stream. These concentrations could easily approach or surpass the concentrations expected during a decontamination using the concentrated or dilute processes. The radiation exposure rates and the associated problems for the mechanical process will be as great, if not greater, than those associated with either of these processes.

THE IMPACT OF DILUTE PROCESS DECONTAMINATION

The majority of dilute process decontamination agents contain weak acids or salts of weak acids, and organic chelating agents such as EDTA or their salts. Other dilute processes entail modification of the oxidation characteristics of the coolant to effect a change in the deposit structure which causes deposited crud to release from the surface.

Materials

Corrosion in decontamination solution is kept low by the use of mild chemicals or inhibitors. Ayres⁶ reports that oxalic acid corrodes carbon steel at a rate of about 0.6 mg/cm²-hr. He also noted that 400 series stainless steels (such as 410, 416, and 420) corrode at about the same rate as carbon steel in common, inhibited decontamination solutions.⁶

In the Can-Decon process⁷ which uses very dilute (0.1 wt% total chemicals) solutions, the Canadians observed quite low corrosion rates, even on carbon steel (see Table I).

The addition of oxygen to the primary system for decontamination will increase corrosion, though exposure time periods are so short that little total corrosion would be expected. In Canadian tests at Douglas Point Reactor⁷ little corrosion was noted even though redox cycling, low pH, thermal shock, and mechanical shock were used (see Table II).

Operations

During dilute decontamination, the corrosion products are removed in a cation exchange column and the chemicals removed by mixed bed ion exchangers following the decontamination.

TABLE I. Corrosion During Can-Decon⁷

<u>Material</u>	<u>Average Penetration Rate, $\mu\text{m/h}$</u>
Carbon Steel	0.28
410 Stainless Steel	0.08
316 Stainless Steel	0.02
Monel-400	0.03
Nickel	0.02

TABLE II. Corrosion of Douglas Point Generating Station Autoclave Coupons⁷

<u>Metal</u>	<u>Total Corrosion μm of Metal</u>	<u>Number of Decontaminations</u>	<u>Exposure EFPD</u>
Carbon Steel	1.3	Zero	209
	1.2	4	239
	1.6	14	341
Monel	0.3	Zero	209
	0.3	4	239
	0.13	14	341
410 Stainless Steel	0.25	Zero	209
	0.3	4	239
	0.5	14	341

The major procedural change during decontamination from normal operation will consist of an increased rate of use of ion exchange resin. This occurs because of the higher removal rate of corrosion products.

The amount of ion exchange resin required for a dilute decontamination is not large. For a 100,000 gallon (3.8×10^5 l) primary system and assuming decontamination with 0.05% oxalic acid plus 0.05% citric acid, there are about 7200 g-equivalents of anions to be removed. According to Clark⁸, the average anion resin has a capacity of 1.2 gram eq/l. Therefore, about 6000 l (210 ft^3) of resin are required for dilute decontamination. Consequently, the equivalent of about three columns are required if a bed size of 100 ft^3 is assumed.

Waste Solidification

The principle processes used for waste solidification are: cement, urea-formaldehyde (UF), asphalt (bitumen), and plastic such as the DOW process. The primary advantage reported for the vinylester is its ability to hold water. The urea-formaldehyde process has been plagued by free-standing water problems.

From discussions with reactor operating personnel, it appears that all solidification systems are difficult to maintain. It is not known whether service people have the same problems or whether because they specialize in one area (waste disposal) they are able to maintain the system in operating condition.

It appears that bitumen and cement are suitable for resin solidification. In addition, discussion with some of the suppliers of various plastic processes suggest that the plastics may be excellent means for solidifying ion exchange resins.

Radiation Exposure

During a reactor decontamination using a dilute process, such as the Can-Decon, radiation exposures in the radwaste treatment system occur as a result of working with or near process resin columns and filters.

As discussed, three 100 ft^3 resin beds for removing corrosion products from the effluent stream are needed. Each of the resin beds will be preceded by a filter for removing particulates. Since the fuel will be in the reactor during this decontamination, we will assume that approximately 10^4 Ci of ^{60}Co will be deposited on the resin and the prefilters. The dilute process effluent will most likely have a low particulate concentration and, therefore, most of the radioactivity will be deposited in the resin beds. For exposure rate calculations, it is assumed that 500 Ci of ^{60}Co is

deposited on each filter and 3000 Ci on each resin bed. The exposure rates on the surface of the resin columns may vary greatly due to concentration distribution in the columns; however, at ten meters nonuniform distribution is not critical (see Table III).

Waste from the dilute process will consist mostly of spent resin and filter cartridges. The resin and filters may be shipped in a cask or placed in 55-gallon drums. The resin and filters may also be combined into a single package for shipment. The spent resin may be mixed with a solidification agent or dewatered and packaged without a binder.⁹ The amount of ^{60}Co in a shipping cask (150 ft³) may vary from about 2700 Ci to almost 5000 Ci. The amount of ^{60}Co in a 55-gallon drum filled with resin may vary from about 130 to 220 Ci.

Table IV lists the exposure rate for radwaste from dilute process decontamination and from typical spent resin from operating plants. These dose rates indicate that a remote facility is probably necessary to work with unshielded spent resin beds. During the site visits it was evident that in most cases adequate shielding was not in place. This indicates that design should include more shielding for decontaminations.

Regeneration of the spent resin, though technically feasible, is being replaced by single usage followed by disposal for normal waste.⁹ The same procedure is anticipated for decontamination cleanup resin. The high exposure rates from spent resin used in the dilute process decontamination make the single use an extremely attractive way to save occupational dose.

TABLE III. Radiation Exposure Rates from Unshielded Resin Beds Containing 3000 Ci of ^{60}Co

<u>Location From Resin Bed</u>	<u>Resin Evenly Distributed on 100 ft³ Bed</u>	<u>Resin Concentrated On 8 ft³ of 100 ft³ bed</u>
Surface	590	4800
1 m	140	180
10 m	3.5	3.5
50 m	0.13	0.13

TABLE IV. Exposure Rates for Spent Resin in
55-Gallon Drums or 150 Cubic Foot Casks

<u>Waste Type</u>	<u>Container</u>	<u>Typical Surface (R/h) Exposure Rate</u>	<u>Average</u>
Decontamination	Drum	210-900	
Decontamination	Cask	400-1300	
PWR	Drum	13-56	
PWR	Cask	0.5-1000 ¹⁰	615 ¹⁰
BWR	Drum	6.2-27	
BWR	Cask	0.33-200 ¹⁰	90 ¹⁰

Radiation exposure will also be encountered from handling the spent filter cartridges. If each filter contains about 500 Ci of ⁶⁰Co, the exposure rates at one meter will be approximately 200 R/h. This makes remote handling of these filters a necessity. Approximately two feet of concrete is needed to reduce exposure rates to the level where workers could be in the general area.

Filter cartridges from operating PWR's are typically placed in 55-gallon drums or small liners for disposal and then placed inside a shielded container. Radiation exposure rates near unshielded filter cartridge packages range from 0.1 to 40 R/h with an average reading of 6.5 R/h.¹⁰ According to Mullarkey, et al¹⁰, occupational exposure resulting from handling the used filter cartridges is high due to high exposure rates. Since filters used during decontamination are likely to have exposure rates on the order of 5 to 2000 times the normal value, remote handling will be required.

THE IMPACT OF CONCENTRATED PROCESS DECONTAMINATION

Concentrated decontamination consists of the application of one or more solutions, generally of 5-10% concentration, to remove activated corrosion products and/or rupture debris from reactor primary systems. The three chemical agents that have been most frequently used, or are scheduled for use are phosphoric acid, AP-Citrox*, and NS-1 (Dow Chemical Company proprietary solution).

*A two step process using alkaline permanganate as the first step and citric acid and oxalic acid and their salts as the second step.

These processes require extensive rinsing to remove the chemicals from the primary system between steps and before restoring normal coolant chemistry conditions.

Materials

Phosphoric acid. From data gathered for this report, it is clear that hot inhibited phosphoric acid should not be allowed to remain in the reactor for extended periods. At ambient temperature, however, the corrosion rate of various stainless steels in uninhibited phosphoric is less than 25 $\mu\text{m}/\text{yr}$.¹¹

The effect of inhibited phosphoric acid on carbon steel is shown in Table V. It can be a fast, effective decontamination solution for carbon steel. Because of its corrosiveness, storage of uninhibited phosphoric acid in contact with carbon steel cannot be recommended, even at normal room temperature.

AP-citrox. The alkaline permanganate (AP) step is required for decontamination in certain systems, such as those containing stainless steel and having a reducing environment.

Ayres⁶ notes that AP is not very corrosive to stainless steel (300 or 400 series) or to carbon steel. The corrosion rate of carbon steel at 105-110°C is about 6 $\mu\text{m}/\text{day}$ which is well below the 60 $\mu\text{m}/\text{day}$ listed by Ayres as a maximum recommended for decontamination.

TABLE V. Carbon Steel Corrosion Rates^{12, 13}
In Phosphoric Acid ($\mu\text{m}/\text{d}$)

<u>Material</u>	<u>Average Corrosion</u>
ASTM A-216 WKA	41.6
ASTM A-105 GR II	41.2
SAE 4140	58.3
ASTM A-515 GR 70	39.0
ASTM A-615	51.7
ASTM A-234 GRWP II	67.4
ASTM A-1603	65.2
Time of exposure	15-30 min.
Temperature	67-82°
H ₃ PO ₄ concentration	7-10%

There are several areas of concern with AP.⁶ First, chromium plate and hard-facing alloys are attacked. The attack on chromium occurs both by direct dissolution and by attack at the chromium/substrate interface. The hard-facing alloys, such as Stellite suffer dendritic attack which is similar to intergranular attack. Water flushing is recommended to protect these surfaces when in contact with AP.

Another problem associated with AP is that any residual may cause caustic cracking when the system returns to temperature.⁶ Also alkaline permanganate is an excellent oxidizing medium and as a consequence, must not contact organic components.

The Citrox step of the AP-Citrox process can be used alone or as the individual citrate or oxalate components. Citrox or its components alone are not highly effective on stainless steel in PWR's though they are reported to be in BWR's due to increased oxidation state. The data for ammonium citrate shows that carbon steel corrodes at rates as high as 20 mg/cm²-hr (25 µm/hr). The use of inhibitors can greatly reduce this (see Table VI).

Other data show that:

- In inhibited oxalic acid, carbon steel has a corrosion rate of 6 µm/day; aluminum, 18 µm/day; and Zircaloy-2, 7.4 µm/day.
- In Citrox, both 1010 carbon steel and 440C stainless steel corrode at a rate of 120 µm/day while the corrosion rates of 300 series stainless steel and Zircaloy-2 are undetectable.
- Titanium is corroded by solutions containing oxalate.

The solutions for the above results are probably 4-10% at about 85°C although this is not reported. Exposure times are on the order of hours.

During a four-hour decontamination of a fueling machine, Ontario-Hydro observed the corrosion rates given in Table VII.¹⁴ This decontamination was performed using a 6% solution of Decon Turco 4512A (an inhibited proprietary citrate-oxalate mixture) at 85°C.

Though the corrosion rates in AP-Citrox solutions are relatively high, the chemicals are not kept in the reactor long and, consequently, the total corrosion is not high.

TABLE VI. Effects of Inhibitors in Dibasic Ammonium Citrate Solutions^(a) on Corrosion Rates of Carbon Steel⁶

Inhibitor	Concentration (g/liter)	Average Corrosion (mg/cm ² -hr)	
		60°C	85°C
None	---	.14	5.1
Phenylthiourea	5	0.2	1.0
Acridine	1	0.2	0.7

NS-1. Results of the test showed that for 1020 carbon steel, some low alloy steels (2-1/4 Cr-1 Mo) and the 400 series stainless steels, the corrosion rates were about 25 to 125 μm per year.¹⁵ The 300 series stainless steels yielded general corrosion rates of less than 125 μm per year.¹⁵ Copper, nickel, chromium, and specialty alloys (such as Hastelloy and Stellite) gave, generally, similar or lower corrosion rates.¹⁵ The effect of temperature in some cases was significant. Incoloy 800 at 250°F had corrosion rates of 175 μm per year whereas at 275°F, the reported rates were ten times as large.

Significant effort was also placed on studying stress corrosion cracking. The study showed that Fe^{+3} or Cr^{+6} in concentrations greater than 0.12 wt% were required to cause intergranular stress corrosion cracking in descaled sensitized 304 stainless steel.¹⁵ Cracking also occurred with sensitized, scaled, 304 stainless steel in the same solutions without the added iron or chromium.

TABLE VII. Corrosion Data from Component Decontamination Using Inhibited Citrate-Oxalate Mixture.¹⁴

Material	Corrosion Rate ($\mu\text{m}/\text{d}$)
304 Stainless Steel	0.6
410 Stainless Steel	2.4
Aluminum Bronze	6
17-4 PH	10.8
Tungsten Carbide	6
Nickel	8.4
Chrome Plating	0.6

Operations

Preparation for decontamination like any other new process entails a significant amount of documentation. Considering the present widespread opinion of the relative time loss associated with concentrated process decontamination versus dilute process, considerably more documentation is expected to be required to justify the use of the concentrated process.

Additional internal documentation will be required for procedures, both operating and emergency. These may well include special communication lines as well as temporary lines of authority. After the decontamination, people will need to know how the operation was conducted, mistakes made, and its effectiveness.

With the present state-of-the-art, a concentrated process decontamination requires complete removal of the fuel, its temporary storage and then refueling. If done as an extension of a normal refueling, the added time may not be excessive.

A special step at least in magnitude, is the preparation of the decontamination chemicals. In the dilute process which might involve only a ton of chemicals, the preparation can be done in a small vessel to make a concentrate which can be pumped into the reactor. In the case of the concentrated process, on the order of 50 tons of chemicals will be required. Preparation and temporary storage of this quantity of chemicals will be required.

Waste Solidification

Phosphoric acid. Phosphoric acid, in 8-10% concentration, is about one molar acid and is, therefore, strongly acidic. It can be solidified by mixing with an absorbant (such as Zonolite) and cement. There are also other proprietary processes used.

AP-citrox. The most probable action with waste AP-Citrox is to mix both chemicals together which neutralizes them and reduces the number of waste tanks needed for storage. The order of mixing is important to prevent possible evolution of ammonia. In addition to neutralizing the chemicals, the mixing reduces permanganate to Manganese Dioxide. As MnO_2 precipitates, it removes much of the radioactivity from the liquid.

No difficulties are anticipated in mixing this product material with cement. The use of bitumen, however, can be hazardous due to the presence of manganese dioxide, a strong oxidant.

Chemical similarity of urea formaldehyde and vinylester suggests that the plastic processes will be acceptable for AP-Citrox waste.

NS-1. No information has been obtained on the compatibility of NS-1 with bitumen. In various discussions it was found that NS-1 is not compatible with urea formaldehyde or cement and is one reason that the Dow vinylester system was developed.

Liquid Storage Capacity

For concentrated processes decontamination, the volume requirement is large for liquids. At all times during a decontamination there needs to be at least one system volume of rinse water available to remove the corrosive chemicals from the equipment.

For a single step decontamination, a minimum of two system volumes would be required with larger amounts of storage depending on the system and desired procedures. For two step decontamination processes, a minimum of six system volumes storage capacity is needed.

Radiation Exposure

During a reactor decontamination using a concentrated process, such as that proposed for Dresden-1, radiation exposures in the radwaste treatment system occur as a result of work associated with maintenance and with the concentration, solidification, storage and/or shipment of the used decontamination solution and the contaminated rinse water. Occupational exposures may result from working near evaporators, pumps, waste solidification equipment, storage tanks for used decontamination solutions, evaporator concentrate, and transporting or storing the solidified waste.

Some aspects of this process do not lend themselves to comparison with normal radwaste system operations. For example, the amount of liquid storage needed during a concentrated decontamination far exceeds that needed during normal operation or that found at currently operating reactors.

If, as indicated by our survey of reactor operators, there are solidification facilities that are not operating, a means of solidifying waste must either be brought in or built on-site. In our contacts with utilities, it was mentioned that once the ^{60}Co is removed from the primary system, the reactor operator may need a license amendment so that he may keep and handle the waste.

The amount of liquid storage needed for a one-step decontamination process and first rinse is approximately 200,000 gallons. Assuming that the amount of decontamination solution and rinse water for the first rinse is about equal to one primary system volume, then there will be approximately 100,000 gallons of each. At least one additional rinse may be necessary which raises the total liquid used to 300,000 gallons. If the decontamination solution and rinse water are directed to separate tanks, there will be a larger amount of activity in the tank containing the decontamination fluid.

Some holdup of corrosion products can be expected in the radwaste treatment system. If the system is new, a rapid buildup of radioactivity could occur. A study done by Babcock and Wilcox found that radiation fields increased steadily during initial stages of operation in which only normal solutions were processed. This indicates deposition of activated corrosion products as the major source of activity in the evaporator.² The same study also found that radioactive material² collected preferentially in the system. The pumps for the evaporators, however, were not major sources of radiation fields. Decontamination solutions are expected to behave similarly.

Contamination in the form of deposits, scale and sludge forms on the evaporator shell walls, heat exchanger tube bundles and bottom well. The major portion of the exposure results from these deposits.²

Plateout or deposition of corrosion products is of concern during any maintenance which may be required during or following treatment of decontamination waste. For normal radwaste system operations, the deposited corrosion products in evaporators, tanks and piping contribute almost all the radiation field near these components. In some cases, the deposited corrosion products may even result in a higher exposure rate when the system is drained. If this also holds true for decontamination wastes, it may be impossible to do any maintenance in some cases and necessitate removal of equipment by remote operations before maintenance can begin. For example, pumps are not expected to accumulate much radioactivity and therefore will not be great sources of radiation and could be maintained with hands-on operations. However, if pumps are located in close proximity to waste evaporators, bends in piping or deadlegs, they may need to be removed from the system before any maintenance can be conducted. Such factors must be considered in design of radwaste treatment systems for decontamination wastes.

Radiation exposure rates near piping in the radwaste treatment system may be five or six orders of magnitude greater for the

decontamination waste than for normal waste (see Table VIII). Shielding of piping is, therefore, an important consideration in the design of a radwaste system which is to handle decontamination wastes. Additional exposure rate problems may occur as a result of plateout or deposition of corrosion products in low-velocity flow areas such as instrument lines, near bends in the pipe, or low points in the system or deadlegs.

Once the waste has been processed through the evaporator, it will be solidified. Urea-formaldehyde might be avoided as the solidification agent due to residual water problems and its incompatibility with decontamination solutions (notably Dow's NS-1). Cement, asphalt or a plastic will in all probability be used.

The evaporator concentrate is placed in 55-gallon drums and solidified. Radiation exposure calculations for the concentrated process are based on removal of 6000 Ci of ^{60}Co . However, it is more likely that the waste will not be thoroughly mixed. Those drums which contain the solidified waste from the decontamination fluid could contain as much as 5000 Ci of ^{60}Co total. Since this radioactivity is contained in approximately one-third the liquid to be solidified, there would be 350 drums with about 15 Ci of ^{60}Co in each drum.

During normal reactor operations the specific activity of solidified waste is much lower than that encountered during a decontamination.

TABLE VIII. Exposure Rates (R/h) Around
Twelve Inch Pipe

<u>Distance</u>	<u>No Shielding</u>		<u>1 Ft. Concrete Shielding</u>	
	<u>Decon*</u>	<u>Normal**</u>	<u>Decon*</u>	<u>Normal**</u>
Surface	5	1×10^{-4}	4×10^{-2}	7×10^{-7}
1 m	5×10^{-1}	1×10^{-5}	8×10^{-3}	2×10^{-7}
10 m	1×10^{-2}	3×10^{-7}	2×10^{-4}	4×10^{-9}

*Decon waste has ~ 10 $\mu\text{C/cc}$ of ^{60}Co .

**Normal waste has $\sim 6.6 \times 10^{-4}$ Ci/cc of mixed fission products and corrosion products.¹⁶

Exposure rates for solidified wastes are listed in Table IX. As seen from the table, exposure rates associated with decontamination waste are about two orders of magnitude greater than the average exposure rates from normally generated wastes.

Radiation exposure rates that are this much greater than ordinarily encountered may lead to problems not only for handling the waste, but for shipment and/or disposal of the waste. Mullarkey, et al, state that, at the high end of the specific activity range, as much as two inches of lead shielding is required for transporting the waste.¹⁰ For decontamination wastes, even more shielding may be required.

One other waste problem is associated largely with two concentrated process. During the decontamination operation, there will be some leaks in the primary and the radwaste system due to action by the solution. The liquid from these leaks are best caught on some sort of absorbing media so that concrete in the containment vessel does not absorb the solvent. This absorbed solvent must somehow be removed from the containment vessel and entered in the waste process stream. Significant radiation exposure of workers could result during this operation.

COST CONSIDERATIONS

The cost of preparing for decontamination is dependent on many factors that have no firm value and on a few factors that are relatively easily defined. Some of these factors are discussed in this section.

TABLE IX. Exposure Rates From Solidified Waste in 55-Gallon Drums

<u>Description of Waste</u>	<u>Exposure Rate (R/h)</u> Surface	
Decontamination waste with 6 Ci of ⁶⁰ Co per Drum	10	
Decontamination Waste with 15 Ci of ⁶⁰ Co per Drum	25	
Normal Solidified Radwaste	0.00005 to 1 ¹⁰	(Average ¹⁰ 0.185)

Organizational Considerations

When considering a decontamination effort, the reason for the decontamination must be defined. If it is a major need, a new or enlarged facility may be required and justifiable. If it is a relatively small job, perhaps a modification to the existing rad-waste facility can be made or the decontamination waste can be handled on a contract basis by a service company.

The decision should be made as to whether the facility will be used repeatedly or only once. This will help define the economics of decontamination.

Two questions, which should be strictly technical and, therefore, relatively straightforward to answer, are: "What decontamination process should be used?" and "How will the final waste be disposed of?" Due to the changing state-of-the-art in processing and to rapid changes in disposal alternatives, these questions become heavily dependent on personal and specific organizational opinions.

Licensing

The areas of licensing and environmental impact must also be considered. The degree to which these topics must be addressed will significantly affect the cost of decontamination. Discussions with some of the contacts suggested licensing costs can range from 25 to 100% of the facility cost. No licensing costs are included in the estimates given below.

Design

The estimates provided here are preconceptual estimates. These are sometimes referred to as scoping estimates. These estimates are typically prepared on the basis of dollars/ft². The \$/ft² figures are obtained from recently completed, similar nuclear construction projects. These data are available from the Department of Energy. By using data from recent projects that are complete or nearly complete, contingency is included. Escalation is not included in these estimates.

Dollar/ft² data are typically presented in the form of ranges. Considering that the facilities and equipment described in this report would have to meet the appropriate seismic and tornado criteria in the Nuclear Regulatory Guides, the high end of the range was selected. The figure is about \$650/ft².

Mechanical Decontamination

Mechanical processes are used on a localized basis and, therefore, will generate a relatively small quantity of waste. It is assumed that existing tanks will be used; these may require additional shielding.

Because the size of equipment decontaminated and the amount of contamination is unclear, no effort is made to estimate costs of facilities or shielding.

Dilute Process Decontamination

The reference radwaste system is assumed to have two ion exchange columns, each with 100 ft³ of resin. It has been estimated that three ion exchange columns are needed for system cleanup plus one for chemical regeneration during decontamination.

With only limited capacity available in the existing facility, and to keep it from becoming badly contaminated, it is assumed that a small shielded facility will be constructed adjacent to the existing radwaste facility. The new facility will contain room for four columns plus associated pumps and filters. The new facility will also contain storage space for hot radioactive resin. The existing gaseous radwaste system is assumed to be used.

Two concepts are considered -- a conventional, hands-on maintenance facility, as shown in Figure 1, and a facility designed for remote* operation, as shown in Figure 2.

Each option considered consists of two floors with the main operating equipment on the main floor. The second floor has support facilities including HVAC and chemical make-up.

For a "standard" hands-on, four-column operation, such as in Figure 1, the estimated facility cost, including process equipment, is about \$2.6M. In addition, as estimated, \$0.8M is required for containment penetrations and \$0.5M for in-reactor piping.

The remote facility, as shown in Figure 2, which would be expected to have lower radiation exposure costs, has a higher capital cost of about \$4.4M. Additional costs are for containment penetrations and piping.

* As used here, remote means that there are no hands-on steps in either normal operation or maintenance operations.

AREA = 2332

FIGURE X. Four Column Hands-On Radwaste Facility -- Dilute Process

FIRST FLOOR = $34 \times 128 + 14 \times 34 = 4828$

SECOND FLOOR = 4352

²
FIGURE 2. Four Column Remote Radwaste Facility -- Dilute Process

Concentrated Process Decontamination

The present, reference, radwaste system is assumed not to meet the needs for concentrated decontamination for reasons, as discussed above, concerning materials, throughput, storage constraints, and shielding. It is assumed, therefore, that a new facility will be constructed. This new facility will contain six tanks (each 100,000 gallons), an evaporator, and associated pumps. Space is allocated for solidification and drumming; costs include estimates for the equipment, although this may be left to a contract service. It is assumed that the existing gaseous radwaste system will be used.

Both a conventional, hands-on maintenance facility and a remote facility are estimated (see Figure 3 and Figure 4). The hands-on facility is a schematic made from drawings of the new Dresden-1 radwaste system. Using \$650/square foot as a facility cost, the Dresden facility is estimated at about \$29M which is close enough to the \$28M reported in our informal industry survey to ensure some confidence in the footage cost.

The remote facility, as shown in Figure 4, is based on the Dresden facility but with a revised layout. This facility is estimated to cost about \$38M.

The cost of an add-on facility is the only cost considered. The cost of including the needed facility in the original reactor construction is not estimated but it would clearly be less. In an original construction, duplication could be reduced and a more optimum use of walls and space could be made.

SAFETY CONSIDERATIONS

In this subsection, factors concerned with safety that would arise from the use of decontamination are considered.

The safety discussion is presented in such a manner that analysis of the opportunities and constraints for using decontamination are fully determined and assessed. The treatment of safety should lead to questions for reviews and to assist utilities and other organizations considering decontamination in assessing what the ramifications might be.

General Considerations

The information presented on general considerations will emphasize concepts and provide a "roadmap" to consider the impact

24A

FIGURE 12.3 Radwaste Facility at Dresden-1 (15)

243

⁹
 FIGURE 12A. Remote Radwaste Facility -- Concentrated Process

of decontamination on safety. OSHA and state requirements will control and direct these types of analyses and the conditions that should be imposed upon such operations. A general statement of the considerations involved and their impact if decontamination were used on a reactor system are given so that a full assessment of impacts can be made.

Chemicals and Chemical Toxicity

When chemical decontamination of a reactor primary system is undertaken, sizable quantities of chemicals are involved. That means that operations, be they for the dilute or concentrated processes, shift the operation of the nuclear facility from power generation to chemical processing. The operators at nuclear reactor systems are trained to handle the mechanical, electrical, health physics and nuclear tasks necessary to generate electric power. The supporting personnel are trained to help these technical people accomplish their tasks. Whenever appreciable amounts of chemicals are introduced such as occurs in decontamination, chemical processing and other backgrounds are required. This shift in background needs and experience has often led conventional and nuclear power utilities to choose chemical service organizations to undertake these operations. When that is done, the service personnel have to be familiarized with site conditions, procedures and operations, in addition to the mandatory constraints commonly encountered at nuclear sites.

Whenever such a safety assessment is made there is a need to involve knowledgeable safety professionals. Industrial hygienists will be able to assess what the major impact will be on personnel from the chemicals that may be used for decontamination. They will also be able to determine if special toxicity measurements are required such as were undertaken at Dresden.¹⁵

Fire Prevention

The primary emphasis on fire prevention techniques is expected to be focused on chemicals and chemical operations that are a part of a decontamination operation. The decontamination materials or their reaction products that are discharged to the radwaste system are potential explosion and fire hazards. The Manual of Hazardous Chemical Reactions¹⁷ can be used to obtain information about these effects.

Fire prevention has another facet insofar as decontamination and its impact on radwaste treatment are concerned. The most significant impact of temporary or modified facilities and equipment is on the existing fire prevention procedures at the plant undergoing decontamination. Because there are unusual or off-standard operations, access to buildings and facilities may be blocked or impeded. A professional fire prevention specialist should be employed to assess the effects of decontamination and the operation of the radwaste system relative to fire prevention procedures.

Working Conditions

Decontamination, because it is not a routine operation, will have a significant safety effect on working conditions. Some of these ramifications have to do with the type and expertise of the personnel needed to undertake the decontamination operation. Other aspects have to do with the stress factors involved with the personnel operating the facility under off-standard conditions.

It should be recognized that decontamination is an off-standard operation and this means that all of the routines and approaches to reactor and radwaste treatment system operation will have to be rethought. Changes lead to different operational conditions and the staff must be fully cognizant about how to respond. Detailed operating procedures must be prepared for and approved for these new conditions. A very detailed training program is needed to teach the operational crews to follow approved procedures. Otherwise, whenever some event occurs, the tendency for the operators is to fall back on standard procedures and the thinking that is used for routine operations.

Probably the most important effect on working conditions is that which will result from the stress on personnel. Any kind of sustained campaign operationally will place stress on the personnel if they are required to function in a new and unusual manner or are retained at work for prolonged periods of time. The work of Morgan, et al.,¹⁰ showing the difference between performance and capacity as a function of successive periods of continuous work is illustrated in Figure 5. We recommend that any decontamination operation have at least two different crews to undertake the operation.

FIGURE 5. Impact of Successive Periods of Work on Work Errors (18)

The place where redundancy is most likely to be overlooked is with the decontamination expert or the leader of the decontamination campaign. This person can become fatigued. At least two people should be counted upon to provide direction for these off-standard operations. Onsite rest is a poor substitute; the specialist is always available and someone awakens him often.

Weather conditions must also be considered. Weather can have a significant impact if part of the facilities where the chemicals are mixed and fed into the system are located out of doors. Personnel comfort and facility integrity are important considerations in severe or inclement weather. In an assessment of the overall safety that is involved in decontamination, due consideration should be given to these conditions.

Organization

It is especially important that off-standard operations have organizational structures that clearly show lines of authority and areas of responsibility. Because different types

of people, observations, risks and procedures are encountered, revised organizational structures are needed to ensure that safe conditions are maintained. Regular reactor and radwaste operating maintenance crews should be included in such an organization. Others, such as the chemical operators involved in undertaking the decontamination and the leaders of that effort, and any others involved in these special operations must also be included into a formalized structure. It is a matter of defining clearly who is going to do what under what conditions.

Specific Considerations

A generic discussion of special problems is presented with a focus on those aspects that might be encountered during the decontamination of a nuclear reactor and the handling of the waste from that kind of an operation. This treatment is not inclusive but is intended to focus on the implications of generic problems. It can best be used to formulate questions about safety ramifications in specific areas of concern.

Mechanical Decontamination

One of the most important problems arising from mechanical decontamination is the possible reaction of metal particles that are generated during such processes with acidic solutions in the radwaste treatment system. Such reactions result in the generation of hydrogen.

Design of these facilities must be done in such a way that it can accommodate hydrogen evolution if they have contact acid solution or explosion and fire problems will ensure.

Another safety impact on the radwaste treatment system is the activation of metal particles that are left in the primary system. These will become extremely radioactive as they pass through the core, will find their way to low spots in the system, and may find their way into the radwaste system when mechanical manipulations of the primary system dislodge them. These highly radioactive particles will likely enter the radwaste treatment system after the decontamination has been completed. They will cause much higher radiation levels than would normally be encountered or expected.

Dilute Process Decontamination

The primary safety concerns for dilute processes will be those having to do with chemicals and chemical toxicity, but specific care

should be addressed to chelating compounds and oxalic acid that are frequently used in such processes. Chelating agents such as EDTA, if ingested, are known to strip calcium from the body causing specific physiological problems. The ingestion of oxalic acid is very dangerous leading to the precipitation of calcium in the kidneys, causing irreparable damage to the body. These examples are given to show what can be involved in this kind of an operation.

Concentrated Process Decontamination

Probably the most important safety problems for concentrated chemical decontamination comes from the large amounts of chemicals that are used in such operations. The 40 tons or more that are expected for such operations must be properly stored, mixed, introduced into the reactor system, removed from that system, introduced into the radwaste treatment system and processed for disposal in a safe and acceptable manner. Some processes may generate sizable amounts of ammonia gas. This gas must be accommodated in the design of the system or arrangements made to control the production of ammonia gas.

WASTE DISPOSAL ALTERNATIVES

Handling wastes generated by decontamination is similar for any of the processes which have been discussed. The impact on disposal of any method of decontamination is very similar. Perhaps the only significant differences are the waste volumes generated and the radioactive content of the resultant concentrates.

There are currently only three operating disposal sites. Any of these sites would be capable of disposing of wastes from decontamination. Their licenses allow them to possess from 50,000 to 60,000 curies of radioactive material at any one time. There are no license or corporate restrictions on size or weight of material which they may handle.

All sites are permitted by their license to bury solidified waste and, at least two of the sites, cannot accept waste which contains any liquid.

Other than these restrictions and some applying to special nuclear material which is not a consideration for normal decontamination operations, the restrictions are based on being physically able to handle the waste. The disposal companies want to keep occupational exposures as low as reasonably achievable and assure

that offsite doses remain well below existing standards. If a shipment exceeds the normal capacity of the disposal operation in terms of size, weight or exposure rate, increased operational costs will be passed on to the customer for any special equipment or procedure which must be used.

It does not seem likely that there is any great obstacle to disposing of decontamination wastes at these sites given the current conditions. There is an indication, however, that the states in which the disposal sites are located could establish more stringent guidelines regarding radioactive waste disposal. How this situation would impact disposal of decontamination wastes must await decisions by the state governments.

One alternative to immediate disposal of decontamination wastes is to store the waste onsite. Temporary storage onsite is not entirely a new concept. All waste must be kept onsite for some time before it is shipped to a disposal site and, at Surry Power Station, steam generators containing many curies of ^{60}Co are being stored onsite until the plant is decommissioned.¹⁹

Licensing problems may exist, but these can probably be overcome. The major question is one of economics. There are several distinct advantages to onsite storage:

- Fewer shipments since radioactive decay allows either higher concentrations or more drums or both per shipment.
- Less radiation exposure at the final disposal site.
- Lower disposal costs due to reduced radiation levels.

An analysis of the potential radiation dose to an individual residing about 500 m from the onsite storage building was done. Buildings with one foot of concrete shielding and with two feet of shielding were considered. No additional shielding from housing or geologic effects were considered. If the radwastes were stored onsite for 25 years, the integrated dose to this individual would be about 700 mrem with one foot of shielding and 15 mrem with two feet of shielding. The maximum annual dose to this individual would be 96 mrem and 2 mrem for the one foot and two feet of shielding, respectively.

If waste is stored onsite the amount of shielding used should be sufficient to ensure that doses are within regulatory limits for the site. Location of the storage building onsite would have to be such that normal traffic flow (pedestrian and motorized) would not pass near the structure with great frequency. There is little off-site dose from this option, even considering a large inventory of ^{60}Co , and storage onsite should not be restricted for this reason.

The particular option for waste disposal that a specific site chooses depends on the site location with respect to the populace, regulations affecting the site, the total inventory to be stored, availability of burial space, costs for the various options, land available onsite for a storage building, and many other factors. The important issue is that there be a safe and reasonably economic way to eventually dispose of the waste permanently.

CONCLUSIONS

The Pacific Northwest Laboratory study of the influence of decontamination on LWR radwaste treatment systems produced several key conclusions and recommendations. These are listed below:

1. Only at N-Reactor is there a means to accommodate radwaste produced during decontamination. The Dresden system is expected to be ready to accommodate such solutions by the summer of 1979. Other radwaste treatment systems at power reactors would require extensive modifications to accommodate radwaste from decontamination.
2. Solidification of the processed decontamination waste may be a significant problem. Solidification agents may not be compatible with the chemicals in decontamination wastes. Urea formaldehyde has residual water problems. Cement increases the total waste volume and weight. Asphalt has potential stability problems.
3. The disposal of the large amounts of concentrated wastes resulting from the operation of the radwaste treatment system may be a problem. Such waste has to be handled and stored onsite, transported and ultimately disposed of in an acceptable site.

4. Based on discussions with utilities, there is doubt that the materials of construction in current radwaste treatment systems are capable of handling the chemicals from a concentrated process.
5. The total storage volume, for concentrated decontamination, is not sufficient in existing radwaste treatment systems. It is also clear that the ion exchange systems are neither large enough nor properly shielded to handle highly radioactive dilute process waste. Current radwaste systems do not have remote operating capability. Such a capability may be required for decontamination wastes.
6. Greater attention should be placed on designing reactors and radwaste treatment systems for decontamination. Such design should consider the proper equipment layout and spacing to accommodate access, maintenance and modifications. In addition, adequate specification of materials and means for introducing and removing chemicals from reactors and radwaste treatment systems must be made.
7. A means of handling waste material resulting from leaks in the primary system during the decontamination must be developed. The radwaste system must be capable of accepting and handling these wastes. Significant radiation exposures could be encountered from handling waste due to system leaks.
8. On-site storage of solidified decontamination wastes may be a viable option, but license amendments will be necessary.
9. To determine the cost incentives for decontamination, long-term benefits associated with reduced occupational exposure and increased operating efficiency should be balanced against the immediate costs for the decontamination, including building modification, reactor downtime and personnel exposure. This is particularly true when cost estimates range from \$3,900,000 for a small hands-on maintenance facility for dilute process waste to upwards of \$38,000,000 for a completely remote operated and maintained facility for concentrated decontamination process waste.

10. Decontamination safety must be addressed. Analysis should consider chemical toxicity, changes in operating procedures, fire prevention and work conditions. Probably the most important effect on working conditions is stress on personnel and human fatigue.

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