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PRE-DESIGN SAFETY ANALYSES OF CESIUM ION-EXCHANGE COMPACT PROCESSING UNIT

W. G. Richmond
M. Y. Ballinger

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Pacific Northwest Laboratory
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William G. Richmond
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The U.S. Department of Energy (DOE) Hanford Site is located in south central Washington state. The 1,456-sq-kilometer (560-sq-mile) site was chosen by the government for the production of plutonium for use in the Manhattan Project during World War II.

Hanford tank wastes began to collect when fuel elements irradiated in Hanford reactors were dissolved for plutonium extraction for nuclear weapons. The waste from these extractions, and a number of other waste-generating projects, has accumulated in underground waste storage tanks on the Hanford site since 1944 (Lang 1993).

Because there are safety concerns associated with continued storage of the complex wastes in the storage tanks, and because some of the tanks have leaked, the need for tank waste remediation has taken priority at the Hanford Site. The processing of radioactive materials in the DOE system has traditionally been conducted in large centralized canyon-remote maintenance facilities. The cost of such new facilities is estimated at approximately $1 billion. This large cost motivated the Pacific Northwest Laboratory (PNL) to develop a new concept for treating Hanford tank waste: a small relocatable process unit termed the compact processing unit (CPU) (Figure 1). The CPU drastically reduces the cost of waste treatment facilities by reducing the need for process support facilities (e.g., HVAC systems), minimizing operator interaction by process automation, and allowing offsite fabrication of the entire process unit. The safety of waste processing activities is improved by the reduced in-process inventory and by process automation.

DESIGN DESCRIPTION

The DOE, through its Office of Technology Development, is funding PNL to develop this innovative radioactive waste pretreatment concept. This cost-effective, highly flexible processing approach is based on the use of CPUs to treat highly radioactive tank wastes in proximity to the tanks themselves. The units will be designed to treat tank wastes at rates from 8 to 20 liters per minute and have the capacity to remove cesium, and ultimately other radionuclides, from 4,000 cubic meters of waste per year. This new concept is being integrated into Hanford's tank farm management plans by a team of PNL and Westinghouse Hanford Company scientists and engineers.

The conventional design philosophy for processing highly radioactive material centered on developments in the early 1940s. This design concept philosophy is called "canyon-remote" design. The basic principle of this design is that all components must be remotely replaceable using an impact wrench and crane. The designs that result from the application of this philosophy rely on simple components and require significant direct operator interaction for successful operation. The level of worker interaction requires a large operating staff within the facility. The presence of the operating staff within the facility in turn requires that the designs include features to minimize worker exposure to radioactive or hazardous materials during both normal operating and accident conditions. The approach used to control worker exposures in these facilities is to incorporate thick shielding walls to protect the workers from radiation and to use ventilation and personnel protective equipment (e.g., respirators) to protect the workers from airborne hazardous materials (chemicals and radioactive contamination). The performance requirements for these systems are very strict due to the direct impact of a failure on worker safety (Boomer 1993).

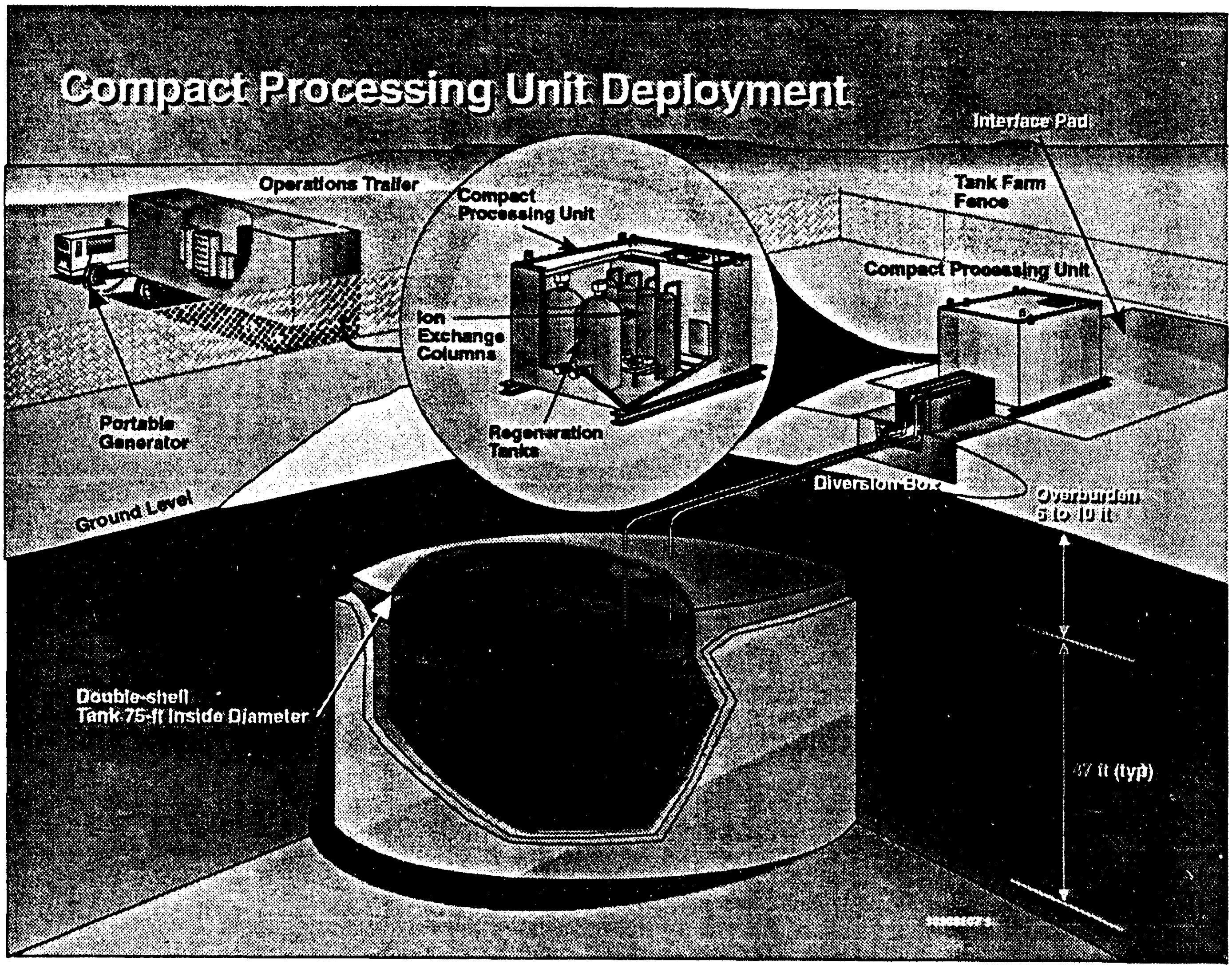

FIGURE 1. Compact Processing Unit Deployment

The CPU design concept departs from this design philosophy by eliminating the need for direct operator interaction and hence eliminating the presence of an operating staff within the facility. In order for this design concept to be practical for use it must be shown that the design can be operated safely without direct operator interaction and in the absence of operator interaction.

The CPU differs in design and operating concepts from the conventional design approach of radioactive material processing facilities. This created the need to verify the viability of the CPU design concept early in the design process. The project coordinators developed safety analysis and risk assessment documentation prior to beginning significant design activities. These analyses determined what project systems structures and components are important to the safety of the onsite worker population and offsite individuals. The analyses also identify key operating procedures that must be incorporated in the design to ensure that safety criteria are satisfied.

The key design features required to meet safety criteria are called safety class systems, structures, and components. The key operating procedure requirements identified are used to ensure that systems must operate under accident conditions. The results of the analyses identify, at an early stage, the design requirements for systems to maintain their function under such conditions. This allows the designer to limit the number of systems required to withstand accident stresses and to properly identify design criteria for systems that are important to safety. The analyses performed for the CPU showed that the safety class systems were the containment structure and emergency vent system. The analyses also identified that the ability to isolate the containment structure as a safety class function. This resulted in classification of the containment isolation system as a safety class system.

The first CPU to be designed and deployed will be used to remove cesium from Hanford double-shell tank (DST) supernatant waste. Separating Cs from the waste would be a major step toward lowering the radioactivity in the bulk of the waste, allowing it to be disposed of as a low-level solid waste form (e.g., grout), while concentrating the more highly radioactive material for processing as high-level solid waste.

The Cs ion-exchange CPU is designed to meet the following objectives:

- transportable to a site at a Hanford tank farm
- able to process about 4 million liters of Hanford DST supernatant waste in 1 yr with a Cs decontamination factor (DF) sufficient to reduce the waste concentration to less than the U.S. Nuclear Regulatory Commission (NRC) Class A maximum concentration (1 Ci/m^3 Cs) for radioactive wastes (10 CFR 61)
- having a minimum operating life of 1 yr, with minimal maintenance or replacement of Cs processing equipment
- operable in compliance with federal, state, and Department of Energy (DOE) regulations and orders.

The ion exchange system is designed to minimize secondary waste generation by neutralizing only the high cesium concentrated eluant to be sent back to the tank farm. The low cesium concentrated eluant will be stored in a tank within the CPU module to be recycled with fresh nitric acid for future elutions. This will maximize the cesium concentration in the eluant while minimizing generations. Minimization efforts also include neutralization of acidic cesium eluant with incoming basic-side feed streams rather than with fresh sodium hydroxide, resulting in volume reductions for the sodium hydroxide required for processing.

DOE SAFETY CLASS SYSTEMS

DOE nuclear operations require safety analysis reports (SARs), technical safety requirements (TSRs), and evaluation of unreviewed safety questions (USQs) according to the 5480 series of DOE orders. This series of orders applies to nuclear operations requirements for nonreactor operations.

Consequently, PNL SARs for nuclear facilities will include an analysis of chemicals in the facility only as they have potential to impact the radiological source terms. Frequently, this evaluation will examine the two most important aspects of large radiological and large chemical sources in the same facility: 1) the potential for chemicals to provide an energy source for the release of radionuclides and 2) the potential for a chemical release within the facility to incapacitate operations personnel such that a radiological hazard would be created or the consequences of a radiological incident would be increased.

The DOE standard for safety analysis techniques establishes guidance for the review of hazard analysis and accident analysis. Order 5480.20, *Nuclear Safety Analysis Reports*, states requirements for DOE nuclear facility safety analyses, hazard classification parameters, and the compliance submittals required to meet the DOE Order. The direction of the Order is to provide guidance and a "graded" approach to assessing SAR compliance. The requirements indicated within DOE Order 5480.20 indicate the path taken by PNL researchers to assess the safety needs and potential accident scenarios a CPU may encounter.

APPLICATION TO Cs CPU

The CPU containment system encloses the process, mitigates process accidents, and shields the operating staff from radiation. The containment structure is designed to meet the Resource Conservation and Recovery Act (RCRA) secondary containment requirements, DOE and NRC requirements for primary containment, and DOE/NRC requirements for onsite shipping casks for radioactive materials. The enclosure will be constructed of steel/concrete composite and is expected to have walls about 1 meter thick for radiation shielding.

The analysis of accidents for the CPU is simple because no personnel are present in the CPU process module. All operating personnel are located in a separate operations control trailer that is located remotely from he CPU. This makes it possible to analyze the impacts to operating personnel using classical safety analysis techniques. These analyses address both the radioactive material hazard and the toxic chemical release hazard. The radiological hazard was evaluated by comparison with established guidelines for effective dose equivalents. The toxic chemical release hazard was evaluated using Occupational Safety and Health Administration chemical exposure guidelines (threshold limit values) for worker exposures.

Potential accidents that could cause radioactive or toxic releases have been identified and are shown in Table 1. This table includes a description of the accident, an estimate of its expected frequency of occurrence, potential consequences, and prevention/control devices to prevent or mitigate the accident. Four accidents are considered bounding (a module drop, loss of cooling water/chiller, a seismic event, and an ion exchange column explosion) and were selected for further analysis. Within the table, frequency is defined as being either anticipated (A), unlikely (U), and extremely unlikely (EU).

TABLE 1. Preliminary Hazards Analysis for Cs Ion-Exchange Compact Processing Unit

Accident	Hazardous Element	Frequency	Consequence	Prevention/Control
Pipe Leak/Break Inside Process Module	Radionuclide, chemical	A	Liquid fills process module at a rate of 5 GPM until detected; containment system prevents release to environment	Control system monitors system pressure, flow rates, and sump levels and upon detection shuts down the pump to stop the flow of waste into the module
Fire Outside Module	Radionuclide, chemical	A	Loss of electrical power, module over temperature	Hanford Fire Department
Module Dropped After Operation	Radionuclide, chemical	A	Tanks, ion-exchange columns rupture and release contents to containment; module remains sealed	Rigging load tested prior to use, containment designed to withstand drop from maximum lift height
Loss of Cooling Water/Chiller	Radionuclide, chemical	A	Liquid in module boils after 24 hours, activating vents on ion-exchange columns	Emergency electrical power
Site Blackout	Radionuclide, chemical	EU	Processing stops, all valves fail closed and seal process module	Backup electrical power, fail-safe vales to seal containment
Ion-Exchange Column Explosion	Radionuclide, chemical	EU	Ion-exchange columns release media to containment	Administrative controls to prevent improper operation of the ion-exchange columns
HEPA Filter Breach	Radionuclide, chemical	EU	Control system detect failure and seals module; temporary exhauster is attached to module in 48 hours	Fire stops on HEPA inlets; back-up connect for temporary exhauster
Electrical Power Failure (primary)	Radionuclide, chemical	U	Process brought to safe shut-down on emergency power	Emergency power system for process shutdown and ventilation system
Fire in Module	Radionuclide chemical	U	Process inoperable and process instrumentation compromised;	Flammable loading limited in process module: combustion air limited in process module
Tornado, Hurricane, High Winds	Radionuclide, chemical	U	Loss of electrical power	Containment designed to with-stand tornado loading
Seismic Event	Radionuclide, chemical	U	Tanks and columns containing liquid waste and ion-exchange resin break and release contents to containment; containment system remains intact and functional.	Containment structure remains intact and contains waste; piping interconnecting process module and tank farm is sealed by valves activated by seismic detector
Loss of Control Center	Radionuclide, chemical	U	Processing stops, all valves fail closed and seal process module	Fail-safe valves to seal containment

Module Drop

The module is assumed to be dropped from a sufficient height so that the impact ruptures the tanks and ion-exchange columns release the maximum inventory inside the module. The module is assumed to remain intact (requiring design of the module outer containment to withstand an impact from the maximum credible drop height). The module is assumed to be agitated enough to support a fog-like concentration of droplets (10 mg/m^3) or a maximum quasi-stable aerosol concentration of fine particles (100 mg/m^3), resulting in the evaporation of fog (Mishima, Schwendiman, and Ayer 1978). The 100 mg/m^3

concentration is bounding and is used in the analysis. The aerosol is assumed to all be in the respirable range.

The maximum onsite exposure is assumed to be at 100 m. Since the CPU is transportable, this distance is also used for evaluating offsite consequences, as there is some possibility that the module may be located within 100 m of the site boundary. Thus, the estimated dose to a maximum offsite exposure is the same as that to a maximum onsite exposure. Unit dose values from a ground-level release to a 100-m exposure in the 200 East Area has been estimated using the computer code GENII. These values for Cs-134 and Cs-137 are 0.85 and 0.6 rem/Ci, respectively. The dose to the maximum onsite and offsite exposures from this event would be 2.6E-3 rem (1.27E-5 Ci x 0.85 rem/Ci + 4.34E-3 Ci x 0.6 rem/Ci). This value is below the PNL dose guidelines of 0.01 rem effective dose equivalent (EDE) offsite and 0.1 rem EDE onsite for an accident with an "anticipated" annual frequency.

This analysis relies on both the integrity of the outer containment to limit the quantity of aerosol generated and at least one set of high-efficiency particulate air (HEPA) filters to mitigate the release. Failure of either of these elements could result in above-guideline dose consequences for this event.

The toxic consequences from the scenario are also estimated. The estimated worst-case inventory of chemicals in the CPU is shown in Table 2. Chemicals in quantities of 10-lb or less were not analyzed because a 10-lb quantity is considered insignificant to the consequences in the accidents analyzed. To substantiate this screening approach, threshold quantities of highly hazardous chemicals listed in 29 CFR Part 1910 are in quantities of 100 lbs or greater and none of the chemicals in Table 2 are included in this list. Of the 10 chemicals listed in greater than 10-lb quantities, only the following three have threshold limit values (TLVs) and are considered hazardous: NaOH, KOH, and $Na(AlO_2)$. Thus, only these three chemicals are included in the analysis.

Except for nitric acid, the concentration of toxic chemicals at the onsite and offsite locations from a module drop were obtained by assuming the aerosol was composed of droplets from the column solution because this solution has the greatest inventory of nonradioactive toxic materials per gram of aerosol generated. The chemicals were assumed to be released from the module over a period of 15 minutes and no credit was taken for filtration. The 95 percentile E/Q for a ground level release in the 200 East Area to a 100 m point is 6.0E-2 s/m^3. This value was determined by the GENII program using the 9-year average meteorological data for the Hanford area. The size of the CPU module does not justify use of a building wake model, so no credit was taken for potential building wake.

Table 3 shows the concentration of toxic chemicals 100 m from the accident site. Table 3 also shows the TLVs for most of the chemicals of concern. These values are normally used to indicate guideline exposures in the workplace and are used in this analysis because more appropriate acute toxicity values are not available. The 100-m concentrations shown in Table 3 are below the TLVs for each of the chemicals and, thus, are very conservatively shown to be below a level that would cause adverse health effects.

The consequences of this accident are also well below the draft ERPG-1 concentration. Since the toxic release from this scenario results in concentrations of toxic materials well below workplace exposure limits and ERPG-1 concentrations the toxic materials present in the CPU do not significantly effect the safety classification of the CPU. In addition, since the concentration of hazardous materials is below the TLV values, the chemical release from this accident scenario will not impact the ability of the operating staff to safely operate the CPU. This analysis shows that from a toxic release standpoint only the containment boundary would be classified as a safety class system. This distinction may allow the design and surveillance of the HEPA filters to be a radiological concern only, which may allow for simpler RCRA permitting of the CPU.

Loss of Cooling Water/Chiller

Cooling water for chilling capabilities is provided to the CPU to remove radioactive decay heat. Without cooling capabilities, solutions inside the tank could boil. With a maximum inventory, solution boiling would not be expected to occur for almost two weeks because of the large amount of solution associated with the inventory. Cesium supplies most of the heat. Energy from the other radionuclides is insignificant because of the low inventories. Heat generation rates are 4.02E-3 W/Ci for Cs-137 and 1.02E-2 W/Ci for Cs-134. Consequently, with a maximum inventory, as much as 4,940 watts could be generated.

Under worst-case conditions, boiling would not be expected to occur for days, leaving adequate time for restoration of cooling before solutions started to boil. However, preboiling solutions may generate airborne releases. Studies examining this release mechanism (Mishima, Schwendiman, and Radasch 1968) found that only minute amounts (0.001 wt% maximum) of plutonium were released during the slow heating of nitrate solutions. Cesium is considered a semi-volatile and may produce a higher release fraction. However, at the low heat generation rate expected, only small amounts of cesium would be emitted. Mishima and Schwendiman (1973) studied releases of cesium under burning conditions and found that low concentrations (0.2 ppm) in a flammable liquid released 0.25% into the air when the liquid was burned. Slow heating of a contaminated solution is not expected to generate as much aerosol as burning a contaminated flammable liquid.

Toxic consequences are expected to be less severe in this scenario than for the module drop because the metals would not be driven off in the slow heating conditions. Very minor amounts of chemicals could become airborne but would not be greater than that from a module that was vigorously shaken in a drop and, since these consequences were acceptable, no further analysis of toxic releases was conducted for this accident scenario.

Seismic Event

A design basis earthquake for the Hanford 200 Area is 0.2 g (Kaiser Engineers Hanford Company 1991). This level corresponds to an annual frequency of about 1.8E-4, which is in the "unlikely" annual frequency category. This earthquake corresponds roughly to a 6.4 on the Richter scale and the following descriptions of damage would be expected to occur: panel walls thrown out of frames; fall of walls, monuments, chimneys; sand and mud ejected; drivers of autos disturbed.

TABLE 2. Inventory of Non-Radioactive Materials

Measured Constituent	Inventory (mols)	Assumed Chemical Form	Molecular Weight (g/mol)	Inventory (lbs)[1]
Water	134520.5	H_2O	18	5333
Nitrate	12303.4	$NaNO_3$	85	2303
Hydroxide	18056.3	NaOH	40	1591
Nitrite	7700.3	$NaNO_2$	69	1170
Aluminum	3677.5	$Na(AlO_2)$	82	664
Potassium	3802.1	KOH	58	486
Carbonate	730.5	Na_2CO_3	106	171
Chloride	519.8	NaCl	58	66
Phosphate	79.0	Na_3PO_4	164	29
Sulfate	38.2	Na_2SO_4	142	12
Iron	27.7	$Fe(OH)_3$	110	7
Chromium	11.0	$Cr(NO_3)_3$	238	6
Ammonia	51.6	NH_4O_H	35	4
Lead	5.2	$Pb(NO_3)_2$	331	4
Zinc	17.2	$Zn(OH)_2$	99	4
Uranium	3.3	$UO_2(NO_3)_2$	394	3
Magnesium	7.7	$Mg(NO_3)_2$	148	3
Silicon	15.5	SiO_2	60	2
Bismuth	2.1	$Bi(NO_3)_3$	394	2
Zirconium	2.0	$Zr(NO_3)_4$	339	1
Fluoride	14.3	NaF	42	1
Calcium	2.9	$Ca(NO_3)_2$	164	1
Molybdenum	2.1	$Mo(NO_3)_2$	220	1
Manganese	1.7	$Mn(NO_3)_2$	179	1
Copper	1.4	$Cu(NO_3)_2$	187	1
Silver	1.1	$AgNO_3$	169	0
Cyanide	3.7	NaCN	49	0
Titanium	0.4	$Ti(NO_3)_3$	234	0
Barium	0.2	$Ba(NO_3)_2$	261	0
Cadmium	0.0	$Cd(NO_3)_2$	236	0
Mercury	0.0	$Hg(NO_3)_2$	331	0
Selenium	0.0	$Se(NO_3)_2$	203	0
Arsenic	0.0	$As(NO_3)_3$	261	0
Sodium	35635.6	Included in Other Compounds		

[1] Rounded to nearest pound.

TABLE 3. Toxic Effects from Drop of Cs Ion-Exchange CPU

Chemical	Concentration (g/L)	100 m Conc (mg/m^3)	Threshold Limit Value (mg/m^3)	DRAFT ERPG-1 (mg/m^3)
NaOH	202	0.087	2 Ceiling[(1)]	
$Na(AlO_2)$	85	0.036		
AlO_2	61	0.026	10 TWA[(2)]	
KOH	62	0.027	2 Ceiling	
HNO_3/NO_X	63	0.042		4

[1]Ceiling values are defined as the concentration that should not be exceeded during any part of the working exposure.
[2]TWA is the Time-Weighted Average concentration to which nearly all workers may be repeatedly exposed (8 hr/day, 40 hr/week) without adverse effect. The draft ERPG-1 value for HNO_3/NO_X is also shown. This value corresponds to the maximum airborne concentration below which it is believed that nearly all individuals could be exposed for up to 1 hr without experiencing other than mild transient adverse health effects or perceiving a clearly defined objectionable odor.

The outer module containment and HEPA filters are designed to withstand the seismic event and remain functional. Equipment inside may become shaken and ruptured. If the seismic event occurred during processing, a line through which waste solution is being pumped could rupture, ejecting solution at a pressure of up to 70 psig.

The consequences from this event are about the same as that from the module drop. A dose of 2.6E-3 rem EDE to the maximum onsite and offsite exposure is estimated. This value can be compared to PNL guidelines of 0.5 rem EDE offsite and 5 rem EDE onsite. As with the module drop scenario, the HEPA filters are needed to mitigate the dose so that consequences are less than guideline values. Based on the similarity of the radiological impacts of this accident scenario to those for the module drop, no further analysis of toxic chemical consequences from this scenario was conducted.

Ion-Exchange Column Explosion

The explosion of ion-exchange columns is a safety concern when organic ion-exchange resins are used with solutions containing nitric acid. The mechanism for the "explosion" of an ion-exchange column is the generation of gasses and steam from the rapid self-sustaining exothermic decomposition of the ion-exchange resin contained in the column. This rapid generation may result in pressurization of the ion-exchange column and subsequent failure or "explosion."

The conditions necessary for the rapid self-sustaining decomposition of ion-exchange resins have been investigated as a result of accidents of this type in the DOE complex and incidents in commercial industry. These investigations have shown that for these accidents to occur four things are necessary: relatively high concentrations of nitric acid [>2 mol/liter],sufficient temperature to initiate reaction, poor heat transfer from the ion-exchange column, and undersized or missing pressure relief systems.

The CPU will reduce the probability of this accident by the following measures:

- administratively controlling the concentration of nitric acid supplied to the CPU to the minimum concentration necessary for resin regeneration and verifying this concentration by analysis prior to transfer of the nitric acid solution into the CPU
- providing control system interlocks to prevent transfer of regeneration solutions to the CPU that exceed a maximum safe temperature
- monitoring the temperature of the ion-exchange column and in the event safe temperatures are exceeded, automatically stopping acid addition and flushing the column with water
- providing relief vents (rupture plates) on the ion-exchange columns sized to ensure that column design pressures cannot be exceeded.

The outer containment of the module and the HEPA filters are assumed to remain intact and functional during and after the explosion. The source term and dose consequences from the explosion are ten times that of the module drop or 0.026 rem EDE to the maximum onsite and offsite exposures. This value can be compared to PNL guidelines of 25 rem EDE offsite and 100 rem EDE onsite. The CPU HEPA filters and outer containment are needed to keep the consequences in this scenario from exceeding guideline values.

Toxicological consequences from this scenario are also ten times greater than that of the module drop. These consequences remain below TLVs and would not cause adverse health impacts to maximum onsite or offsite exposures or impair the operating staff of the CPU.

CONCLUSION

PNL developed the concept of a compact or modular processing unit in FY 1991. Evaluations focused on the scientific feasibility of designing and constructing CPUs for waste pretreatment. The study concluded that the construction of compact waste pretreatment units is scientifically feasible and that the technology for the design and construction of ion-exchange CPUs is available. The study also concluded that further investigation is required before CPUs could be shown to be practically deployable as a replacement for a centralized pretreatment facility.

In FY 1992, Westinghouse Hanford Company investigated the practicality of using CPUs as an alternative to a centralized pretreatment facility. This investigation consisted of developing the concept to sufficient detail that a cost estimate for deployment could be made. This cost estimate showed that the deployment of a group of CPUs as a replacement for the initial pretreatment module could result in a significant reduction in the capital cost ($300 - $500 million) required to provide organic destruction and cesium removal processes for the remediation of tanks on the safety watch.

In FY 1993, PNL analyzed the safety impact of the compact processing unit design concept. Since this concept was a departure from current practices, a pre-design safety analysis of the CPU design concept was developed. This analysis considered the radiological and toxicological releases from potential accidents involving the CPU module. The safety class systems in the CPU were identified as: the containment module structure, the emergency ventilation system HEPA filter (1 stage), and the containment isolation system. The consideration of hazardous chemical releases did not change or identify any new safety class systems for the CPU.

The analysis showed that through proper design of the process containment system, virtually any commercially designed process system can be deployed for the use with radioactive tank waste. This should increase the range of processes available for deployment of the CPU and enable greater participation by private industry in the treatment of radioactive tank wastes. Based on these analyses, the CPU program is proceeding with the design of the CPU process equipment. The procurement of this equipment using chemical process industry quality assurance procedures, codes, and standards will be initiated in FY 1994. The use of standard chemical industry design codes and standards as opposed to nuclear industry codes and standards will result in significant cost savings to the DOE.

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