Journal of Hazardous Materials, 35 (1993) 427–441
Elsevier Science Publishers B.V., Amsterdam

High-priority Hanford Site radioactive waste storage tank safety issues: An overview

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Abstract

High-activity radioactive waste has been stored in large underground storage tanks at the U.S. Department of Energy's Hanford Site in eastern Washington State since 1944. Since then, more than 227,000 m^3 (60 Mgal) of waste have accumulated in 177 tanks. These caustic wastes consist of many different chemicals. The waste forms include liquids, slurries, salt cakes, and sludges. A number of safety issues have been raised about these wastes, and resolution of these issues is a top priority of the U.S. Department of Energy. A waste tank safety program has been established to resolve these high-priority safety issues. This paper will deal with three of these issues. The issues described are the release of flammable vapors from single- and double-shell tanks, and the existence of organic chemicals and/or ferrocyanide ion-containing fuel-rich mixtures of nitrate and nitrite salts in single-shell tanks. Extensive management controls are employed to ensure that the tanks in question continue to be maintained in a safe manner through issue resolution. In addition, comprehensive monitoring, characterization, and applied and basic research efforts have been initiated to support resolution of issues and to prevent creation of future problems associated with potentially incompatible wastes. The safety efforts will also support actions related to the planned retrieval and disposal of the wastes in these storage tanks. Such efforts will also provide the basis for safe near-future remediation of these tanks on an as-needed basis and will define the envelope of safety to support the disposal of all high-level waste in the Hanford Site tanks.

1. Introduction

High-level radioactive waste has been stored in large underground storage tanks at the U.S. Department of Energy's (DOE) Hanford Site in eastern Washington State since 1944. Approximately 227,000 m^3 (60 Mgal) of waste have accumulated in 177 tanks. These caustic wastes consist of many different chemicals. The waste forms include liquids, slurries, salt cakes, and sludges. A number of safety issues have been raised about these wastes, and resolution of these issues is a top priority of the DOE. This paper describes some of the

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resolutions of safety issues being pursued at the Hanford Site. An overview of the logic underlying the resolution of Hanford Site high-level waste tank safety issues was recently published and is illustrated in Fig. 1 [1].

2. Background

The radioactive waste stored in underground tanks at the Hanford Site has come from various sources: (1) three different plutonium and uranium recovery processes from approximately 100,000 MTU of irradiated fuel, (2) many different radionuclide recovery processes from waste, and (3) multiple miscellaneous sources (e.g., laboratories and reactor decontamination solutions). The neutralized wastes include sodium nitrate/nitrite, sodium hydroxide, sodium aluminate, sodium phosphate, large amounts of organics (previously not included in inventories), and approximately 775×10^{16} Bq (210 MCi) of radionuclides.

The wastes are stored in 149 single-shell tanks (SSTs) and 28 double-shell tanks (DSTs). The SSTs (Fig. 2) consist of a reinforced concrete tank with a carbon steel liner and have capacities ranging from 208 m^3 (55,000 gal) to 3,785 m^3 (1 Mgal). The DSTs (Fig. 3) consist of a carbon steel inner tank within a steel-lined concrete tank; each has a nominal capacity of 3,785 m^3 (1 Mgal). Sixty-six of the older SSTs have leaked or are suspected to have leaked approximately 3,785 m^3 (1 Mgal). No waste has been added to these tanks since 1980, and the drainable and pumpable liquids are being removed so that the remaining waste will be mostly sludge and salt cake containing minimal interstitial liquid. The first DST was placed in service in 1970; none of them have leaked.

3. Safety issues identified

Safety studies and evaluations have been conducted periodically as new waste-producing processes were developed and waste conditions changed. However, the delay of permanent waste disposal, continual pressure of waste generation on limited storage space, and the aging of facilities have resulted in several current safety issues [2].

U.S. Congress also has expressed concern about Hanford Site tank safety in Section 3137 of Public Law 101-510 [3]. Section 3137 specifically directs the Secretary of the DOE to take the following actions:

- Identify those tanks at the Hanford Site that "...may have a serious potential for release of high-level waste due to uncontrolled increases in temperature or pressure...".
- Determine whether "...continuous monitoring to detect a release or excessive temperature or pressure..." at each identified tank is being carried out

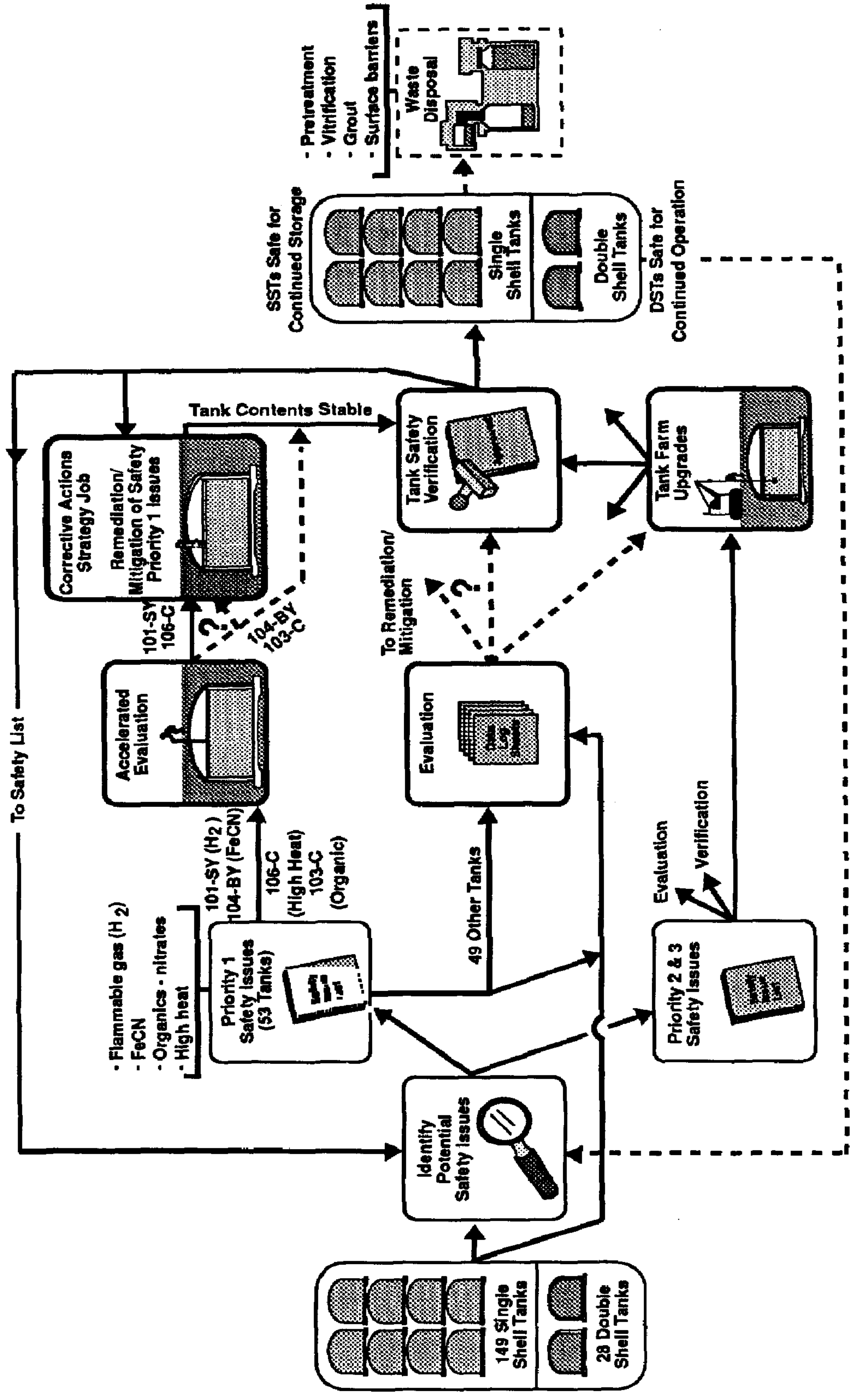

Fig. 1. Potential safety issues.

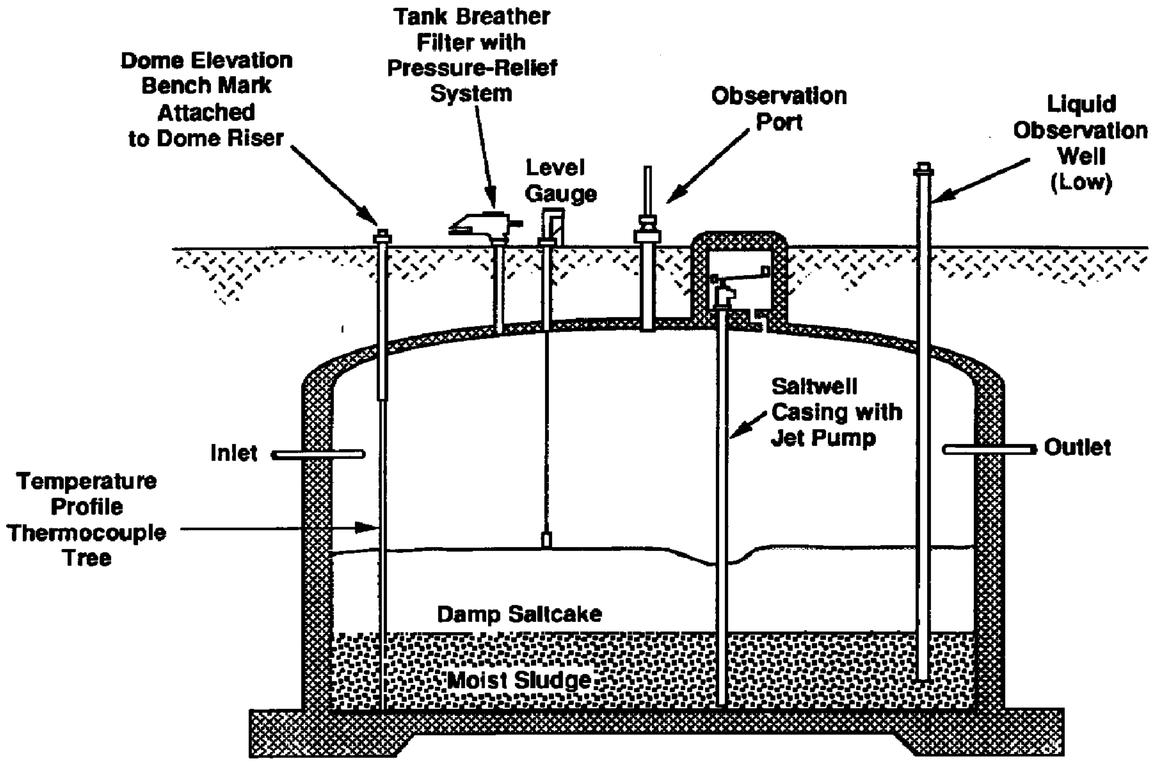

Fig. 2. Typical single-shell tank.

and, if not, install such monitoring as soon as possible if installing such monitoring does not increase the danger of a release.

- "Develop action plans to respond to excessive temperature or pressure or a release from any tank identified..." as having a serious potential for release of high-level waste because of uncontrolled increases in temperature or pressure.
- "Prohibit additions of high-level radioactive wastes to the identified tanks except for small amounts to be removed and returned to a tank for analysis unless the Secretary determines that no safer alternative exists or that the tank does not pose a serious potential for a release of high-level radioactive waste."

As required by Subsection 3137(a), the DOE has identified 53 high-level radioactive waste tanks that "....may have a serious potential for release of high-level waste due to uncontrolled increases in temperature or pressure." Fifty-two of these tanks were identified because they contain chemical materials that, at temperatures much higher than those measured in any Hanford Site tanks, may produce an exothermic reaction. The remaining priority 1 tank has a high ^{90}Sr content and requires periodic water addition for evaporative cooling to maintain tank temperature within specification limits.

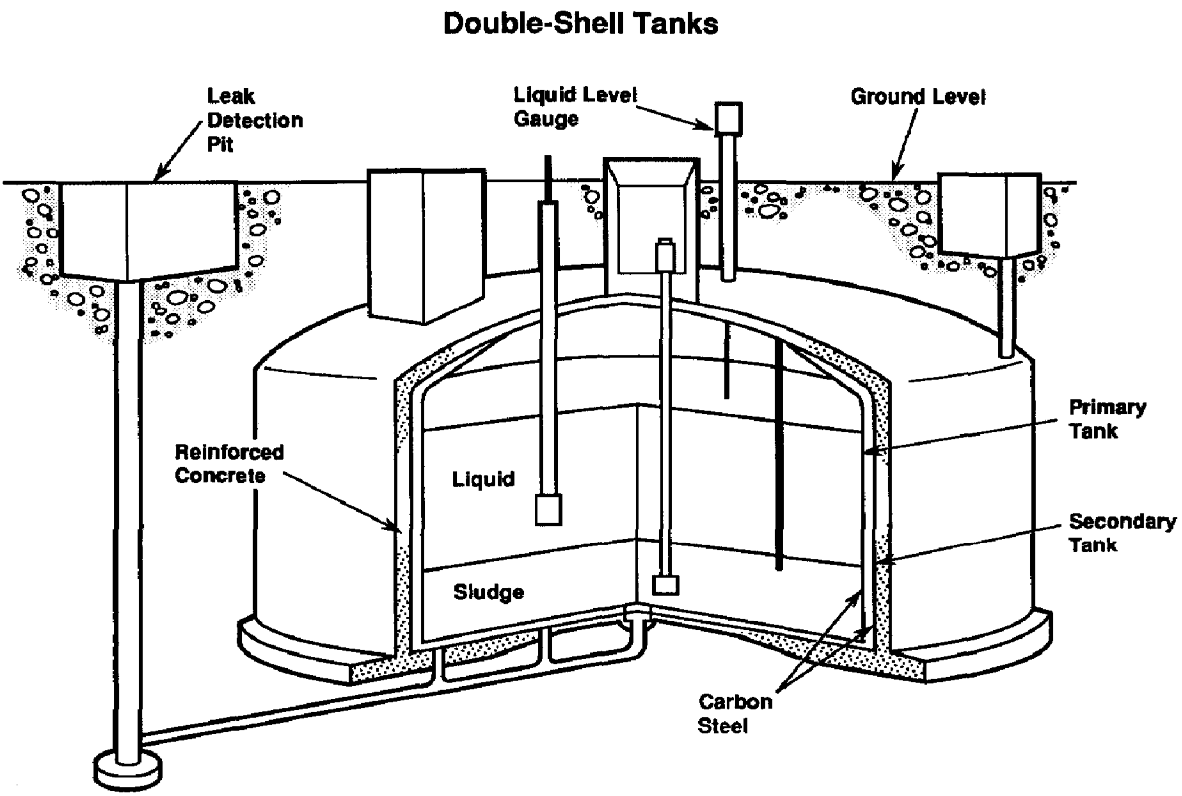

Fig. 3. Typical double-shell tank.

4. Resolution of safety issues

Resolution of all the safety issues will take several years. As the tanks receive further evaluation, it is anticipated that other issues may be identified. A waste tank safety program has been established to conduct this work, and an overview plan for implementing mitigation and/or remediation of waste tank safety issues was prepared (as shown in Fig. 1). Detailed plans also are being developed for each of the major activities.

Issues of concern reported in this paper are as follows:

- Flammable gas generation in Tank 241-SY-101 and other tanks — Twenty-three tanks generate hydrogen and other flammable gases and release them in a periodic episodic fashion.
- Potential explosive mixture of ferrocyanide in tanks — Eighteen tanks contain insoluble ferrocyanide salts in quantities greater than 1,000 moles mixed in a sodium nitrate–sodium nitrite matrix.
- Potential organic–nitrate reactions in tanks — Eight tanks contain organic chemicals at concentrations believed to be greater than 10 mol% sodium acetate equivalent mixed in a sodium nitrate–sodium nitrite matrix. Three of the hydrogen and ferrocyanide tanks also appear on the organic list.

The hazardous characteristics of the existing wastes, leading to their identification and control, were estimated on the basis of general information from the chemical literature, expert peer judgment, and limited historical and actual sampling data. Mitigating factors such as moisture content, presence of inert diluents (e.g., sodium carbonate, sodium aluminate and/or sodium phosphate) and conditions that could lead to a lack of reactivity of the wastes were purposely understated.

Scenarios of significant concern associated with waste in tanks include the following:

- Potential for ignition of flammable gases such as hydrogen–air, hydrogen–nitrous oxide and/or air–organic vapor mixtures.
- Potential for ignition of organic–nitrate and/or ferrocyanide–nitrate mixtures initiated by the radiolytic or chemical heating of dry salt cake or (potentially) by localized heating.
- Potential for secondary ignition of organic–air and/or organic–nitrate mixtures initiated by the burning of flammable gases.

Administrative and technical controls are in place to restrict activities that could cause undesirable exothermic reactions. For example, pumping of interstitial liquid from ferrocyanide tanks has been stopped in order to maintain present moisture levels (e.g., to maintain present thermal conductivity and heat capacities) until more is known about the required moisture levels to maintain safety. Nonsparking tools and electrical bonding techniques are used around hydrogen tanks to prevent accidental ignition. So-called "normal" activities for tanks at issue are limited to surveillance. Special safety analysis documents are prepared for all work inside the tank. These documents are extensively peer reviewed, and work is scheduled for periods when the vapor concentrations are well below the lower flammability limits for hydrogen and nitrous oxide or air.

Of the 53 safety issues tanks, 4 have been selected for accelerated evaluation. Tank 241-SY-101 is of the greatest concern because of episodic releases of flammable gas that exceeded lower flammability limits for short periods of time. Tanks 241-C-109 and 241-C-112 appear to have the largest concentration of ferrocyanide. Tank 241-C-103 has a separable organic layer floating on top of the aqueous waste and is a primary source of potentially flammable vapors.

Although the ferrocyanide and organic tanks are handled, for management convenience, as two separate programs, they both can be classed as fuel-rich tanks.

5. Approaches to ascertaining the risk from fuel-rich or flammable gas tanks

5.1 General considerations

Proof of the conditions in any given tank will rarely be absolute. Rather, the evidence must convince external peer reviewers that the potential for an

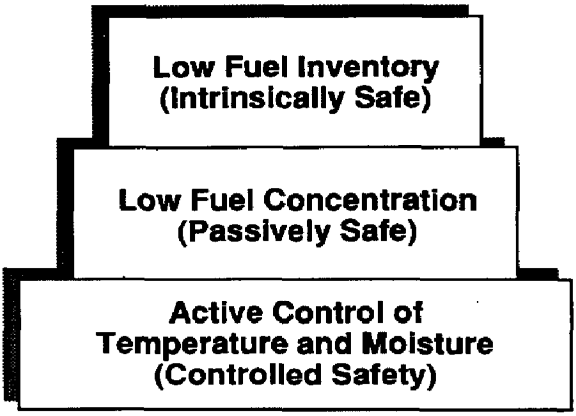

Fig. 4. Safety pyramid.

exothermic reaction in a fuel-rich tank, such as a ferrocyanide salt-containing tank, is very low. Evidence for demonstrating these safety classes (Fig. 4) is not mutually exclusive. Information on the potential safety of a fuel-rich tank can and will be obtained from a variety of activities.

Three long-term criteria affect the evaluation of the safety condition in any given tank. In order to safely store waste in a tank until final disposal is accomplished, the waste must be kept in a safe form. It must be demonstrated on a tank-by-tank basis that the contents of the tank are either intrinsically safe (low fuel inventory), passively safe (low fuel concentration) or in a state of controlled safety (effective monitoring and corrective system in place). Otherwise the contents of that tank must be subject to *in situ* mitigation or early remediation.

Table 1 contains a listing of these factors affecting a judgment of safety and the associated information needed to prove the credibility of the evaluation. However, existing and new data are unlikely to provide absolute proof of safety for any given tank other than those that either received no inventory of fuel or were completely emptied of the suspect materials. Rather, a case will have to be made that the data and analysis results clearly suggest that continued storage of the waste is safe under specified operating safety requirements control.

5.2 Flammable gas tanks

The demonstrated existence of episodically released concentrations of flammable gases in the dome space of any tank above 25% of the lower flammability limit (LFL) creates a situation that automatically makes that tank a candidate for mitigation. The nature of the mitigation process used will depend on tank conditions. As an illustration of this premise, more details on Tank 241-SY-101 are provided in the second half of this paper.

TABLE 1

Information sources associated with evaluation of safety factors

Safety factor	Data source	Information needed
Inventory limit	• Historical information	• Inventory estimates from transfer records • Flow sheet projections
	• Characterization data	• Dome gas sample analysis • Tank sample analysis
	• Modeling	• Dome gas flow models • Transient maximal inventory of flammable gas • Thermodynamic energy estimates
	• Synthetic waste studies	• Gas generation, retention and release mechanisms • Large-scale combustion data (e.g., Bureau of Mines) to define the energetics of a gas burn • Fuel degradation or decomposition
Concentration limit	• Historical information	• Flow sheet analysis • Concentration factors
	• Characterization data	• Multiple point dome gas measurements to define concentration profiles • Liquid and solid tank sample analyses
	• Modeling	• Thermodynamic energy estimates • Thermal modeling of tank responses
	• Synthetic waste studies	• Synthetic waste studies – Fuel dispersion mechanism and/or degradation pathways – Fuel concentration pathways – Initiators and catalysts
Control limits	• Historical information	• Analysis of tank records
	• Characterization data	• Tank vapor sample analysis • Tank surface sample analysis • Concentrations of key constituents
	• Monitoring data	• Enhanced dome space monitoring data • Enhanced moisture and temperature monitoring data
	• Energetics and reaction dynamics	• Modeling the impact of a postulated gas or condensed phase fuel burn • Thermodynamic energy estimates • Thermal and structural modeling of tank

5.3 Fuel-rich tanks

The analyzing of fuel-rich single-shell tanks is more complex. A conclusion allowing safe storage of fuel-rich compounds without modifying the physical position of the waste in the tanks requires a convincing demonstration, on a tank-by-tank basis, that one of the following conditions exists in a high-level waste tank.

- The inventory of organic materials or ferrocyanide salts is below that considered to create a potential for exothermic chemical reaction under any foreseeable tank conditions. Such a demonstration would involve either (1) proving that less fuel than initially estimated was in the tank; OR (2) clearly demonstrating that a mechanism for removal or destruction of fuel under waste tank (inherent safety) conditions exists.
- If the inventory of organic compounds or ferrocyanide salts is sufficient to suggest a risk, that inventory must be shown to be sufficiently dilute (dispersed) so that concentrations necessary for an exothermic reaction do not exist in the tank. This would lead to an assessment of passive safety. Furthermore, it must be proven that no credible mechanism for concentrating fuel to measurements considered hazardous can exist in the tank. These conditions would lead to an assessment of inherent safety.
- If convincing proof cannot be obtained demonstrating that neither of these conditions exists, a case must be made that the safety of the wastes can be guaranteed by a combination of an assured waste moisture content and the fact that no reasonable mechanisms exist for heating the waste (controlled safety). The demonstration of controlled safety conditions in the tank must be supplemented by monitoring key parameters to ensure that (1) the presumed conditions of safety continue for the entire time of storage; and (2) that methods are in place to rapidly mitigate any possible future effects of tank heatup or significant moisture loss.

5.4 Additional considerations

Proof of the conditions in any given tank will rarely be absolute, but demonstrating a safe inventory and/or concentration of fuel in the condensed phase of a million gallon SST poses special difficulties. Rather than "absolute proof", the evidence must convince external peer reviewers that the potential for an exothermic reaction in a fuel-rich SST such as a ferrocyanide salt-containing tank is extremely low. Also, evidence is mounting that maintaining sufficient moisture content to prevent propagation of any exothermic reaction will not be difficult. Information on the potential safety of a fuel-rich tank can and will be obtained from a variety of activities. Evidence for demonstrating that a tank meets a specific safety class is not mutually exclusive. Rather, the data obtained by the program will create a continuum of peer opinion of that tank's safety that will ultimately be integrated by a consensus process into closing the safety issue(s) related to that tank.

The history of the SSTs sheds light on difficulties in dealing with the safety issues associated with them. First, no wastes have been added to any of the SSTs since November 1980. Half of the tanks, 66 of the 149, are classified as assumed leakers. To prevent or diminish impacts of future leaks from SSTs, a program to remove pumpable liquids from these tanks was initiated. After pumping, each tank was isolated from neighboring tanks. A prohibition exists against adding water to SSTs, as a means of preventing leakage to the environment. There is also concern that any attempts to still

TABLE 2

Preliminary guidelines for ensuring safety of flammable gas producing tanks

Safety factor	Characteristic	Criteria	Notes
Inventory limit	None initially applied	• Cyclic tank pressurization • Anomalous increases in tank surface levels	Based on records of tank behavior (past selection criterion)
Concentration limits	Dome gas concentration	Gas concentration >25% of lower flammability limit	Based on industrial fire protection standards (present safety criterion)
Control limits	Dome gas concentration	25% lower flammability limit	Assumes that tank gas concentrations require mitigation to even out cyclic gas releases to below lower flammability limit criterion

TABLE 3

Preliminary guidelines for ensuring safety of fuel-rich tanks

Safety factor	Characteristic	Criteria	Notes
Inventory limit	Total waste quantity	3% total organic carbon or 1,000 mol FeCN salts per tank (present criterion)	Based on initial assessment of risk
Concentration limits	Waste concentration	<100 cal/g (present preliminary guess or estimate)	Specific to actual chemical composition and waste energetics
Control limits			Assumes that data is insufficient to prove inventory or concentration based safety
• Moisture	• Tank moisture content	• 20+% Moisture	
• Temperature	• Tank temperature	• Lack of a fuel concentration mechanism and TBD[a] Btu/hr heat generation • A TBD[a] temperature limit (<85 °C)	

[a]TBD = to be determined.

their contents would increase the risk of a leak, thus further limiting mitigation options. Note that DSTs pose no such intrinsic constraint because their contents can be kept wet and if necessary stirred, as is planned for Tank 241-SY-101.

Examples of criteria that could be associated with a demonstration of safety for flammable gas and fuel-rich tanks are proposed in Tables 2 and 3.

6. Specific issue overview

6.1 Flammable gas tanks

Flammable gas generation in Tank 241-SY-101 is a top priority waste tank safety issue at the Hanford Site because concentrations above the LFL for hydrogen occur periodically in the tank dome space about every 110 days. This periodic release of gases is expected to recur until some form of mitigation or remediation is taken. In the unlikely event that an ignition source were present during these periods, a hydrogen burn or explosion could occur with a possible release of nuclear waste to onsite and offsite personnel. A number of published reports contain technical summaries of recent information of this issue [4, 5].

There are 22 other tanks also suspected of potentially containing smaller accumulations of hydrogen or other flammable gases. There is, however, a significant difference in severity between them and Tank 241-SY-101. Evidence of venting, surface level behavior, and knowledge of these other tank contents suggests a much lower likelihood of potentially dangerous gas concentrations in these other tanks.

The discussions in this section will center on Tank 241-SY-101 because it is currently the focus of efforts to understand and ultimately to remediate the tanks. As Tank 241-SY-101 becomes better understood, attention is shifting toward characterization efforts for Tanks 241-SY-103 and 241-AN-104, which also show periodicity of gas release.

A number of hypotheses have been proposed for the mechanisms of gas generation, retention and release (Fig. 5). However, more information from characterization, modeling, and laboratory simulation studies is needed before one can fully understand and plan to mitigate the tanks. The exact mechanism of gas generation, retention, and release is still not completely defined, and work is continuing at Westinghouse Hanford Company, Argonne National Laboratories, and Pacific Northwest Laboratory to determine it.

Although the probability of a gas ignition event is extremely low, the level of risk from such an event remains unacceptable because of potential consequences. Therefore, after initial but detailed characterization of the contents of Tank 241-SY-101 (and other tanks that undergo cyclic venting), steps to mitigate or remediate the tanks will be evaluated and ultimately implemented.

Demonstrations of mitigation strategies for Tank 241-SY-101 are being developed and will be tested in Tank 241-SY-101. Potential mitigation methods,

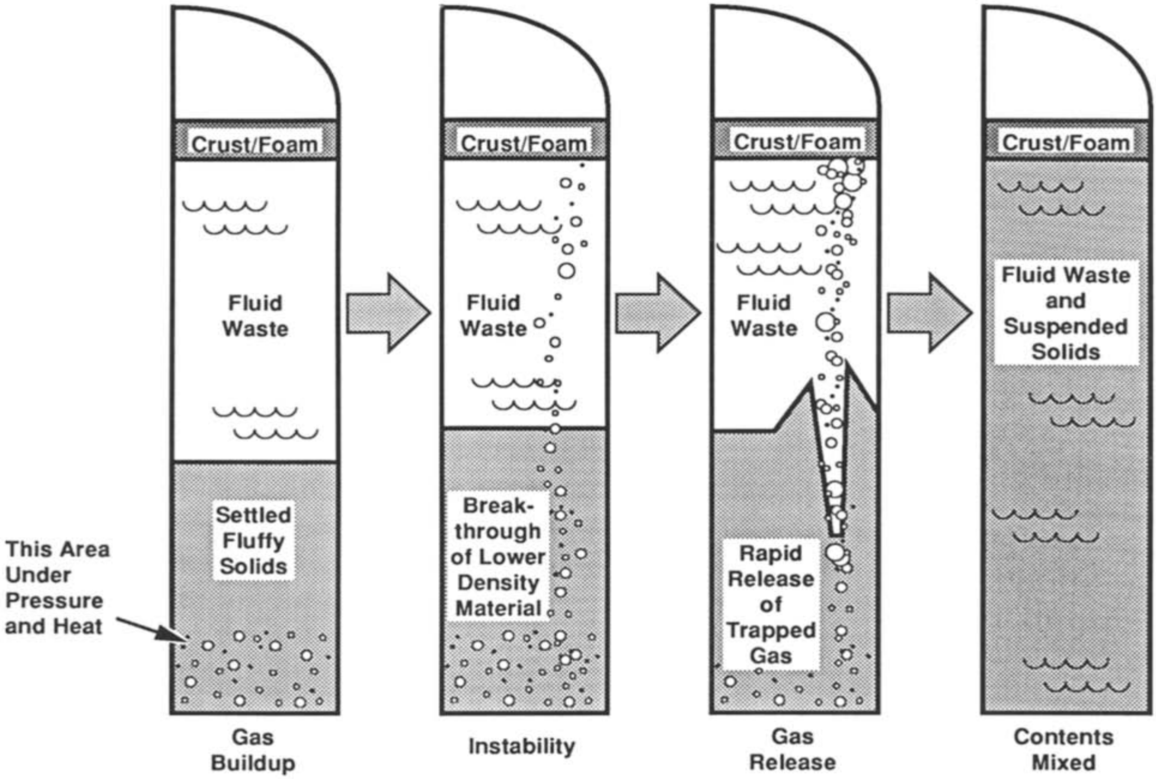

Fig. 5. Postulated mechanism of Tank 241-SY-101 venting.

all aimed at minimizing or eliminating "burp" cycles, include heating and/or dilution of the wastes and/or stirring them to allow the gases that are formed by the chemicals and radionuclides to vent continuously. Other alternatives being explored include ultrasonic methods for forcing continuous release of the gases as they are formed. These alternatives are not mutually exclusive because pumping and use of ultrasound both generate heat in the tank.

6.2 Potentially explosive mixtures of ferrocyanide in tanks

Ferrocyanide tanks were selected as a safety issue because it is not now known whether concentration and distribution of ferrocyanide and nitrate–nitrite materials in the tanks would allow a runaway exothermic reaction if tank contents were allowed to heat up. Although the measured tank temperatures are far below the temperature required to cause an exothermic reaction, the consequences of an event could be at a level potentially exceeding the safety envelope defined in the final Environmental Impact Statement [6]. A number of published reports contain technical summaries of recent information on this issue [7–9].

These tanks store radioactive wastes containing ferrocyanide compounds resulting from the 1950s process used to scavenge radioactive cesium from waste liquids stored in the tanks. To obtain additional storage volume within a short period of time without constructing additional storage tanks, Hanford Site scientists developed a cesium scavenging process to reduce radionuclide levels in tank supernatants to levels that would allow their disposal to cribs. This process involved the carrier precipitation of cesium nickel ferrocyanide along with an excess of sodium nickel ferrocyanide. In implementing this process, up to 140 metric tons of ferrocyanide were added to a group of SSTs.

Concentrated mixtures of ferrocyanide salts in the presence of nitrate and/or nitrite constituents can be made to react and explode under certain conditions. These conditions include dryness, favorable stoichiometry, and elevated temperatures or a high-energy spark. These exothermic reactions can begin at between 180 and 200 °C (356 and 392 °F), and an explosion can occur at 285 °C (545 °F). Maximum temperatures measured inside the ferrocyanide tanks at the Hanford Site are at or below 55 °C (130 °F).

Records at the Hanford Site currently show that there are 18 SSTs that contain appreciable ferrocyanide precipitates (1,000 moles or more). The ferrocyanide content of these tanks ranges from 1,000 g-mol (465 lb) up to approximately 83,000 mol (39,000 lb in Tank 241-BY-104) calculated as the ferrocyanide anion. Other wastes in these tanks probably include significant quantities of sodium nitrate and sodium nitrite and a variety of silicate, aluminate, hydroxide, phosphate, sulfate, carbonate and nitrate salts as well as salts or oxides of uranium, copper, and calcium. In addition, the fission products are also present from the processing of irradiated fuel. Some tanks may also contain quantities of organic materials that cause exothermic reactions to start at the low end of the temperature range listed previously.

The results of synthetic waste tests with materials generated from each of the primary flow sheets used to scavenge radiocesium indicate that about 80% of the ferrocyanide added to the tanks is too dilute in concentration (e.g., diluted with inorganic inert materials) and would be essentially unreactive when heated. The remaining 20% of the ferrocyanide added to the SSTs was put into four C-Farm tanks. The concentration of ferrocyanide in these four tanks is sufficiently high to support a runaway chemical reaction if the mixture were to dry out and become heated significantly above 220 °C (428 °F). However, the ferrocyanide precipitate added to these tanks can apparently retain approximately 50% water, even after centrifugation for 30 gravity years. This estimate is based on simulants prepared in the laboratory using the original 1950s flowsheet. The priority focus of the program is to obtain actual samples from the tanks in C-Farm. Three cores have been obtained from Tank 241-C-112 and are being analyzed for chemical composition as well as characteristics related to their potential energetics.

The probability of a ferrocyanide explosion is considered very low because currently measured maximum temperatures in the ferrocyanide tanks (55 °C [131 °F]) fall significantly below the lowest threshold exothermic temperature (180–200 °C [356–392 °F]) found in the laboratory. Efforts are focused on enhancing monitoring capability, characterizing tanks and gaining information on the mechanism and propagation and radionuclide release characteristics of a ferrocyanide runaway reaction.

A recent review of the practice of pumping out SSTs to avoid the potential for leakage of radioactive and hazardous materials into the soil disclosed that additional analysis of this practice for the ferrocyanide tanks is needed. For tanks that contain large quantities of ignitable materials (ferrocyanide and organic-containing tanks), such salt-well pumping has been discontinued until safety evaluations of liquid removal can be completed.

6.3 Organic tanks

Concentrations of organics may be present in some tanks that could cause an exothermic reaction given a sufficient driving force such as high temperature. However, the difference between ignition temperatures and measured actual tank temperatures, as discussed above for the ferrocyanide tanks, is so large that the probability of such a reaction is considered extremely low. The consequences of the postulated reaction are about the same as the scenarios for an explosion in a "burping" hydrogen or ferrocyanide tank. Although work on this issue is just beginning, consideration of hazards associated with heating nitrate-nitrite mixtures containing organic materials is an integral part of both the hydrogen and ferrocyanide tank efforts.

High concentrations of organic compounds have been inferred from a recent review of tank transfer flow sheet records and limited analytical data in eight SSTs. Many organic chemicals, if present in concentrations above 10 dry wt% (sodium acetate equivalent), have the potential to react with nitrate–nitrite constituents at temperatures above 200 °C (392 °F) in an exothermic manner. The concentrations of organic materials in the listed SSTs and their chemical identity is not accurately known at present. A tank sampling program is being developed to provide more information on the contents of these tanks and to serve as a basis for laboratory testing and safety evaluations.

7. Conclusions

The Hanford Site's Waste Tank Safety Program is large and complex and has high priority within the DOE. Evaluating the safety issues and defining appropriate remedial action to correct these safety concerns is being actively pursued at the highest possible priority. Risk to the operating staff, the Hanford Site environment, and to the general public appears to be extremely low; this is being reaffirmed. Site personnel are working to both quantify the risk and take appropriate corrective actions to support continued safe storage of the waste as well as the eventual permanent disposal of the waste in the Hanford Site's SSTs and DSTs. The road to resolving the safety issues has been mapped out, and although it will be a journey of several more years, the site is well on its way to understanding and resolving these issues.

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