U.S.NRC

United States Nuclear Regulatory Commission

Protecting People and the Environment

Review of Spent Fuel Reprocessing and Associated Accident Phenomena

Office of Nuclear Regulatory Research

AVAILABILITY OF REFERENCE MATERIALS
IN NRC PUBLICATIONS

U.S.NRC

United States Nuclear Regulatory Commission

Protecting People and the Environment

Review of Spent Fuel Reprocessing and Associated Accident Phenomena

Manuscript Completed: October 2016
Date Published: February 2017

Prepared by: N. E. Bixler, F. Gelbard, D. L. Y. Louis and J. Phillips*

*Sandia National Laboratories
 Albuquerque, New Mexico 87185
 Operated for the U.S. Department of Energy

M. Fuhrmann, NRC Project Manager

NRC Job Code Number V6050

Office of Nuclear Regulatory Research

ABSTRACT

The work presented in this report supports the advancement of the safety assessment capabilities of the NRC staff in terms of evaluating accidents that could release radioactivity into the environment and estimating the associated risk during the reprocessing of nuclear fuel. This report lays out much of the background and the concepts needed to construct a computational tool for reprocessing facility source terms. It reviews the following topics: (1) the status of past and current reprocessing plants throughout the world; (2) an overview of a typical plant design, its functions, and its layout; (3) historical accidents and the phenomena that are relevant to these accidents; and (4) the models that are needed to describe these accidents. This document does not propose or describe a specific computational tool that can be used to estimate source terms for a reprocessing facility. Rather, it supports the NRC's capability to evaluate potential accident source terms that would be used in a license application for a reprocessing facility.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

EXECUTIVE SUMMARY

In response to some commercial interests in future reprocessing plant construction and operation, the U.S. Nuclear Regulatory Commission (NRC) is investing in developing expertise to support advanced nuclear fuel reprocessing. The work presented in this report supports the advancement of the safety assessment capabilities of the NRC staff in terms of evaluating accidents that could release radioactivity into the environment and estimating the associated risk.

The history of fuel reprocessing operations in the United States has been limited to the nuclear weapon complex for material production. Licensing of a civilian aqueous reprocessing facility was last sought for the Barnwell Nuclear Fuel Plant about the year 1970. Along with historical reprocessing efforts in the United States, foreign fuel facilities, supporting both weapon development and civilian spent fuel processing, have contributed to the development of various chemical techniques for reprocessing fuel. The co-extraction process is considered to be a candidate for any future domestic civilian aqueous reprocessing without plutonium separation. However, no request for a license has been submitted to date nor has a specific plant design been proposed to be built. Therefore, much of this document focuses on historical plant designs (e.g., Barnwell) to provide reasonable expectations for a future aqueous reprocessing plant.

This report lays out much of the background and the concepts needed to construct a computational tool for aqueous reprocessing facility source terms. It reviews the following topics: (1) the status of past and current reprocessing plants throughout the world; (2) the design, functions, and layout of a typical plant; (3) historical accidents and the phenomena that are relevant to these accidents; and (4) the models that are needed to describe these accidents. This document does not propose or describe a specific computational tool that can be used to estimate source terms for a reprocessing facility.

The various accidents considered are fires, spills, inadvertent criticality events, and explosions. Modeling the relevant phenomena for each accident type is a prerequisite for creating a source-term tool to provide the release rate and aerosol characteristics of a release to the environment. A purpose of this document is to evaluate and describe the relevant phenomena that could contribute to a source term. Later work will focus on developing the source-term tool.

ACKNOWLEDGMENTS

ABBREVIATIONS AND ACRONYMS

AHA	Acetohydroxamic Acid
ALI	Annual Limits on Intake
AMMD	Aerodynamic Mass Mean Diameter
APM	Atelier Pilote Marcoule
ARF	Airborne Release Fraction
ASME	American Society of Mechanical Engineers
AVM	Marcoule Vitrification Facility
BNFP	Barnwell Nuclear Fuel Plant
BRF	Base Recycling Facility
BWR	Boiling Water Reactor
CCD-PEG	Chlorinated-Cobalt Dicarbollide-Polyethylene Glycol
CEMG	Crane and Equipment Maintenance Gallery
CERS	Contact Equipment Removal Station
CFAST	Consolidated model of Fire and Smoke Transport
CFD	Computational Fluid Dynamics
CFM	Cubic Feet per Minute
CRA	Control Room Area
CLS	Cask Loading Station
DAC	Derived Airborne Concentration
DBE	Design Basis Earthquake
DOE	Department of Energy
DOG	Dissolver Off-Gas
DR	Damage Ratio
DTPA	Diethylenetriaminepentaacetic acid
EMS	Equipment Maintenance Station
FDS	Fire Dynamics Simulator
FP	Fission Product
FPIG	Filter Piping and Instrument Gallery
FRSS	Fuel Receiving and Storage Station
GNEP	Global Nuclear Energy Partnership
GP	General Purpose
GSD	Geometric Standard Deviation
HA	High Activity
HAN	Hydroxylamine Nitrate
HAS	Hydroxylamine Sulfate
HEME	High Efficiency Mist Eliminators
HF	Hydrofluoric acid
HILC	High Intermediate Level Cell
HLC	High Level Cell
HLLW	High-Level Liquid Waste
ICPP	Idaho Chemical Processing Plant
ILC	Intermediate Level Cell
ILLW	Intermediate-Level Liquid Waste
INRA	International Nuclear Recycling Alliance
LPF	Leak Path Factor
LWR	Light Water Reactor
MAR	Material At Risk
MOX	Mixed oxide fuel

NPH	Normal Paraffin Hydrocarbon
NRC	Nuclear Regulatory Commission
NUEX	Modified PUREX process
OBE	Operating Basis Earthquake
OML	Oxalate Mother Liquor
PACT	Plasma Arc Centrifuge Treatment
PARE	Pressurized Airborne Release Equipment
PHP	Plasma Hearth Process
PPC	Plutonium Product Cell
PNC	Plutonium Nitrate Cell
PNSA	Plutonium Nitrate Storage Area
PNSL	Plutonium Nitrate Storage and Load-out
PVB	Pressure Vessel Breach
PWR	Pressurized Water Reactor
RF	Respirable Fraction
RMSC	Remote Maintenance and Scrap Cell
RPC	Remote Process Cell
SAC	Sample and Analytical Cell
SNL	Sandia National Laboratories
ST	Source Term
TALSPEAK	Trivalent Actinide-Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes
TBP	Tributyl Phosphate
THM	Ton of Heavy Metal
THORP	Thermal Oxide Reprocessing Plant
TRU	Transuranic
TRUEX	Transuranic Extraction
TVF	Tokai Vitrification Facility
UDS	Undissolved Solids
UN	Uranyl Nitrate
UNH	Uranyl Nitrate Hexahydrate
UPC	Uranium Product Cell
USBM	United States Bureau of Mines
VOG	Vessel Off-Gas
WIP	Waste Immobilization Plant
WTC	Waste Tank Cell
WTEG	Waste Tank Equipment Gallery
WVDP	West Valley Demonstration Project
WVP	Waste Vitrification Plant

1.   INTRODUCTION

In recent years, there has been some domestic interest in reprocessing commercial spent nuclear fuel. The Department of Energy (DOE) is investing resources to study advanced reprocessing technologies and methods. Before a domestic commercial reprocessing facility can be built and operated, it must be licensed by the U.S. Nuclear Regulatory Commission (NRC).

The purpose of this report is to provide the background research and analyses to develop a computational tool to estimate accidental releases of hazardous substances to the environment, or source terms, for fuel reprocessing facilities. Best estimate analyses incorporate state-of-the-art methods, models, and computational codes intended to adequately model and predict the progression of an accident from the onset of the initiating event up to and including characterizing the release of any hazardous material to the environment.

The risk of an accident is defined as the product of the probability and the consequence of the accident. The development of a source term tool should provide a methodology to estimate the consequences of an accident. The probability of an accident is determined separately from the source term and consequence analysis, and may be determined through hazard analysis methods such as failure modes and effects analysis or event tree analysis. Once a determination of the probability of a given event is made, the source term tool estimates the timing and magnitude of potential release to the environment. The resulting source term coupled with a consequence analysis tool, like MACCS, can be used to estimate the distribution of radioactive materials to the surrounding environment and to assess the consequences of the accident.

The various accidents considered are fires, leaks and spills, inadvertent criticality events, and explosions; each of which falls under the purview of the source term tool. Modeling the relevant phenomena for each accident type is a prerequisite to creating a source term tool to provide the release rate and aerosol characteristics of a release to the environment. The purpose of this document is to evaluate and describe the relevant phenomena that could contribute to a source term. Later work will focus on developing the source term tool.

The history of fuel reprocessing operations in the United States has been limited mostly to the nuclear weapon complex for material production. Licensing of a civilian facility was last sought for the Barnwell Nuclear Fuel Plant near the Savannah River Site in South Carolina around the year 1970, while the West Valley reprocessing facility, located near Buffalo New York, operated from 1968 to 1972. Along with historical efforts in the United States, foreign fuel reprocessing facilities, supporting both weapons development and civilian fuel reprocessing, have contributed to the development of various chemical techniques for reprocessing fuel. A brief status of the fuel reprocessing facilities operated to date is presented in Section 2. The co-extraction process is considered to be the most likely candidate for future domestic civilian reprocessing without plutonium separation.

A review of the operations conducted and systems employed at reprocessing facilities is presented in Section 3. The reprocessing stages presented include receiving, storage, separation, and waste management. Brief discussions on the various engineered systems such as ventilation, confinement, and fire sprinkler systems are provided as well.

From the historical operations of fuel reprocessing plants, descriptions of accidents, with some information on offsite consequences, are available. Section 4 presents a history of the accidents categorized by accident type. A general discussion of the relevant phenomena is also presented. A list of these accidents is presented to build familiarization with these events as well as their potential consequences. Of the reported accidents, nitration-oxidation-reaction events comprise the largest category.

Given the occurrence of an accident, an estimate of transport of released radioactive material and ultimate release into the environment is the purpose of a source term tool. Each type of accident and related transport of nuclear materials are controlled by a set of phenomena. The phenomena pertinent to accident progression and transport for the set of accident types are discussed in Section 5.

2. STATUS OF FUEL REPROCESSING PLANTS

Most of the early fuel reprocessing plants in the world were built for military applications. In the U.S., defense fuel reprocessing was performed at several Department of Energy sites, such as Hanford, Idaho, Oak Ridge, and Savannah River. Commercial fuel reprocessing briefly existed in the U.S. until President Carter stopped civilian fuel reprocessing to reduce the possibility of nuclear proliferation.

The following tables give a brief description of defense and civilian fuel reprocessing facilities around the world. Table 2-1 and Table 2-2 summarize the past, current, and planned fuel reprocessing facilities in the world and the current and planned U.S. enrichment and mixed oxide production plants, respectively. Since many modern spent fuel reprocessing facilities include actual fuel production operations and solidification of liquid wastes (vitrification), a list of these facilities is also included in this section. A list of vitrification facilities is presented in Table 2-3.

Table 2-1 World fuel reprocessing plants [GFMR 2010]

Facility	Type**	Operational Status	Capacity (THM/year*)	Notes
Belgium				
Dessel	Civilian	Decommissioned		This reprocessing plant (called "Eurochemic") was built as a joint facility with 12 other OECD states, and was operated between 1957 and 1974. It has been dismantled [IAEA-T1529].
China				
Pilot Plant	Civilian	Starting up	50-100	A long-delayed pilot reprocessing plant at Yumenzhen site in Gansu Province. Another source rated this facility at 0.1 THM/year [NUREG-1909].
Lanzhou [IAEA-T1467]	Civilian		800	A future plan for the high capacity fuel reprocessing plant that is planned to start in 2020.
France				
APM [OCED 2005]		Shut-down in 1997	5	APM (Atelier Pilote Marcoule) is a pilot reprocessing plant built at Marcoule. This plant was commissioned in November 1962 to validate the processes at pilot scale. The actual fuel reprocessing, which began in 1987, was done for several fuel types, including natural uranium-gas-graphite fuel, mixed fuel and fast breeder oxide fuels.
UP1	Dual	Shut-down in 1992	600 [NUREG-1909]	UP1 is located at Marcoule. Large-scale separation of plutonium was carried out for both military and civilian purposes between 1958 and 1997. Reprocessing for military purposes was ceased in 1992.
UP2[A,1]		Operating	1000	The UP2 facility reprocessed gas-graphite reactor fuel from 1966 to 1987.The UP2 plant started reprocessing light water reactor oxide fuel in 1976. In 1994, the UP2 facility was expanded, and renamed UP2-800. One source rated this facility at 850 THM/year [NUREG-1909].

Table 2-1 World fuel reprocessing plants [GFMR 2010]

Facility	Type**	Operational Status	Capacity (THM/year*)	Notes
UP3[A,1]		Operating	1000	UP3 facility is a first major extension at the La Hague site, and started operating in 1989. This facility has been rated to produce 850 THM/year [NUREG-1909].
Germany				
Karlsruhe [A] [NUREG-1909]	Civilian	Decommissioned	25	This facility was operated between 1970 and 1991. [IAEA-T1529].
India				
Trombay	Non-civilian	Operating	50	This facility started operating in 1964 to produce plutonium using the PUREX process. The plant was augmented and restarted operations in 1983 [Dey 2005].
Tarapur	Dual	Operating	100	The original facility was commissioned in 1975 and was intended to reprocess Zircaloy clad oxide spent fuel. This plant uses a chop-leach technique for the head-end and uranous nitrate stabilized by hydrazine as the reductant for partitioning [Dey 2005]. New plant commissioned in 2011.
Kalpakkam	Dual	Operating	100	This facility is intended to reprocess Zircaloy clad natural uranium oxide spent fuel [Dey 2005].
Israel				
Dimona	Non-civilian	Operating	40-100	The fuel reprocessing capability includes a plutonium separation plant (Machon 2).
Italy				
Saluggia	Civilian	Decommissioned	0.1 [NUREG-1909]	This pilot plant was named "EUREX", and operated from 1970 to 1983 [ODEC Italy].
Trisaia	Civilian	Decommissioned		This pilot plant was operated during 1970s to process the uranium-thorium cycle fuels from the U.S. Elk River reactor. It also included a solidification of liquid reprocessed waste (cementation) [OECD Italy].
Japan				
Rokkasho	Civilian	Delayed	800	This facility, which is operated by Japan Nuclear Fuel Limited [Ueda 2005], has design capacity of 8 tons of plutonium or 800 tons of uranium annually. Delayed until at least 2018 A MOX fuel fabrication plant was originally planned to be completed by 2015. This facility remains non-operational [GFMR 2010].
Tokai	Civilian	Temporarily shut down	200	This facility, which is operated by Japan Nuclear Cycle Development Institute, started in 1977 [Ueda 2005].
North Korea				
Yongbyon	Non-civilian	On standby	100-150	
Pakistan				
Nilore	Non-civilian	Operating	20-40	
Chashma	Non-civilian	Under construction	50-100	
Russia				

Table 2-1 World fuel reprocessing plants [GFMR 2010]

Facility	Type**	Operational Status	Capacity (THM/year*)	Notes
RT-1	Dual	Operating	200-400	RT-1 is located at the Mayak site. It reprocesses naval and research-reactor fuel. This plant also separates reactor-grade plutonium and uranium.
Seversk	Dual	To be shut down after cleanup	6000	It contains an isotope separation plant, which became operational in 1953.
Zheleznogorsk	Dual	To be shut down after cleanup	3500	
United Kingdom				
AEA Reprocessing plants [NUREG-1909][C]	Non-civilian	Decommissioned	0.02	There were two plants, one for material test reactors, and one for mixed oxide reactors.
B204[C]	Non-civilian	Shut down		B204 was the first reprocessing plant to extract plutonium for military applications in the UK.
B207	Non-civilian	Decommissioned		This was a uranium purification plant [NUREG-1909].
B205	Civilian	Operating	1500	The B205 Magnox reprocessing plant processes foreign light-water reactor fuel. The original schedule was to shut down this facility in 2012. However due to backlog, shutdown is now planned for 2017.
THORP[A]	Civilian	Temporarily shut down	1200	Thermal Oxide Reprocessing Plant (THORP) at Sellafield reprocesses oxide fuel from its AGR reactors and foreign LWR reactors. Due to many incidents at the plant, THORP throughput varies greatly. Once the current obligations are met, regulators plan to shutdown THORP in 2020 [MSF 2011]. According to [NUREG-1909], there was a miniature pilot plan that was shutdown at THORP.
United States				
Oak Ridge[A] [Manhattan 1999]	Non-civilian	Decommissioned		X-10 was constructed in 1940s. It contained a pilot plant for a chemical separation process to recover plutonium from the experimental pile experiment within X-10.
Hanford [A,B][Bastin 2008], [Manhattan 1999]	Non-civilian	Decommissioned	Total: 96,900 T and B plants: 8100 REDOX plant: 22,400 PUREX plant: 66,400 [Gephart 2003]	Hanford Engineering Works became the production location for producing weapons grade materials during and following World War II. There were three plants – "T", "U" and "B" plants. Because of the large size (60 feet high by 700 feet to 1,100 feet with thick-walled heavily reinforced concrete structures) of the processing equipment, these plants were sometimes referred as "Canyon" [Bastin 2008]. Remotely operated and maintained equipment was installed at the bottom of the structure to carry out the chemical processing. The "T" plant separated plutonium out of irradiated fuel from Hanford production reactors [Manhattan 1999]. The "U" plant was to recover uranium not recovered earlier using a solvent extraction process, and the "B" plant was to recover isotopes from nuclear waste [Bastin 2008]

Table 2-1 World fuel reprocessing plants [GFMR 2010]

Facility	Type**	Operational Status	Capacity (THM/year*)	Notes
Idaho [Pace 2006], [Bastin 2008]	Dual	Shutdown	Over 10 THM during the operating life of the plant [Pace 2006]	Idaho Chemical Processing Plant (ICPP) was configured like the Oak Ridge reprocessing plant. ICPP was built to reprocess all highly enriched uranium irradiated in U.S. production reactors in the 1950s [Bastin 2008]. ICPP was intended to recycle uranium from the plutonium production reactors [Pace 2006]. In 1957, ICPP was an important player for processing non-defense nuclear fuel. All highly enriched fuels including those from the Shipping-port Core were processed at ICPP. Thus ICPP has been reprocessing fuels with cladding in aluminum, zirconium, stainless steel, graphite, and many other unique materials [Pace 2006]. It processed the fuel for space applications and from research reactors as well [Pace 2006]. ICPP was ordered to shut down in 1992.
Savannah River [A] [Savannah 50_CTheseh11], [Bastin 2008], [GFMR 2010]	Dual	H-Canyon is still operating, and the F-Canyon is being decommissioning	15 [GFMR, 2010]	Savannah River Site in 1950s contained two primary fuel reprocessing plants for defense use – 200-F [uses PUREX process] and 200-H (today, they are often referred to as F-Canyon and H-Canyon), which were based on wet processes used to dissolve fuel assemblies and on chemical separation to extract both uranium and plutonium [Savannah50_Ch11]. H-Canyon's current mission is to blend down the highly enriched uranium to low enriched uranium for power reactor fuel applications [GFMR 2010]. Some impure plutonium may be reprocessed in this facility. The extracted plutonium would either be used as feed for the mixed-oxide plant being built at Savannah River Site or vitrified to be disposed as waste.
Morris [West Valley 1996],[NUREG-1909]	Civilian	Never operated	300	This was a General Electric Midwest Fuel Recovery Plant at Morris, Illinois. It was declared inoperable in 1974.
Barnwell [West Valley 1996],[NUREG-1909]	Civilian	Never operated	1500	This facility was constructed in 1970 by Allied General Nuclear Services (AGNS) at Barnwell, South Carolina. This plant was planned to operate in 1974. The plant never operated due to delay in construction and licensing, and the 1977 decision by President Carter to defer indefinitely all reprocessing of commercial nuclear fuel.
West Valley [West Valley 1996]	Civilian	Decommissioned	300	This facility is the first commercial spent fuel reprocessing plant in the US and was operated by Nuclear Fuel Services (NFS) between 1966 and 1972. This facility used the PUREX process. Other operations included in this facility were chopping the assembly rods, dissolving the spent fuel in acid, separating and storing radioactive wastes.

Table 2-1 World fuel reprocessing plants [GFMR 2010]

Facility	Type**	Operational Status	Capacity (THM/year*)	Notes
Energy Solutions [Energy Solutions 2008]	Civilian	Proposed		A Base Recycling Facility (BRF) was proposed for GNEP. BRF is drawn heavily on the THORP Facility at Sellafield in the UK. The facility includes a head-end process facility, where the spent fuel is removed from its transport casks under water, and is placed into temporary storage containers. This facility uses the UREX process method to extract fissile materials.
International Nuclear Recycling Alliance (INRA) [INRA 2008]	Civilian	Proposed	800	INRA proposed a fuel reprocessing plant that is adapted from the technology at the La Hague and Rokkasho reprocessing plants. The plant contains a number of operations: used fuel receipt and storage; shearing, dissolution, and compaction; separations, purification, and concentrations; fuel fabrication; fission products vitrification; high active solid waste interim storage; and waste processing, such as vitrification.

*THM stands for ton of heavy metal.
**Dual means both civilian and non-civilian applications.
[A]PUREX Operation
[B]REDOX/PUREX Operations
[C]BUTEX Operation
[1]The La Hague Reprocessing Site includes two current operated reprocessing plants – UP2-800 and UP3.

Table 2-2 U.S. enrichment and mixed oxide plants [GFMR 2010]

Facility	Type	Operational Status	Capacity * (THM/year)	Comments
Oak Ridge [Manhattan 1999]	Non-civilian	K-25 was decommissioned.		In 1942, a reprocessing pilot plant to be built at Oak Ridge, Tennessee was initiated. K-25 at Oak Ridge was the nation's first diffusion plant for enriched uranium for weapons applications in 1943. This plant used the gaseous diffusion method to enrich uranium. Additional enrichment facilities (such as K-27) were also built near K-25 and the entire complex was called the "Oak Ridge Gaseous Diffusion Plant". The K-25 building was demolished in 2013.
		Y-12 is operational.		Y-12 was built in the1940s and operated in early 1943 for enriching uranium. Y-12 produces uranium with higher enrichment than had been produced at K-25.
Paducah, Kentucky	Civilian	Shut down	11300	Paducah Gaseous Diffusion Plant started the first production cells for enriched uranium in September 1952 to fuel production reactors. The intent of this plant was to produce low-enriched uranium. The low-enriched uranium was then further enriched at K-25. In the 1960s, this plan was converted for producing fuels for commercial nuclear power plants [Paducah 2011].
Eunice, NM	Civilian	Operating	5900	In 2006, Louisiana Energy Service was issued a construction license to build Urenco, a European consortium of enrichment facilities. This facility is based on a gas centrifuge technology to enrich uranium fuel for power reactor use. This facility is the first enrichment plant since 1956.
Savannah River, S.C. [NUREG-1767]	Civilian	Under construction	3.5 (Pu)	Department of Energy's Mixed (MOX) Fuel Fabrication Facility at Savannah River Site has been under construction since 2007. This facility was scheduled to be completed in 2019, but may be canceled.
Piketon, Ohio	Civilian	Under construction	3800	USEC American Centrifuge Plant was first built as a pilot plant. Once the Department of Energy grants the loan guarantee, then the full-scale plant will be built.
Areva Eagle Rock, Idaho	Civilian	Planned	3300-6600	Areva planned to begin construction in 2011 but the project is on hold.
GLE, Wilmington, NC	Civilian	Planned	3500-6000	Global Laser Enrichment (GLE) delayed again a decision on building a commercial-scale facility.

*THM stands for ton of heavy metal.

Table 2-3 Status of high-level waste vitrification facilities [Jain 1998, DOE 1999]

Site	Location	Technology*	Started	Status
Marcoule Vitrification Facility (AVM)	Marcoule, France	Induction melting	1978	Facility shut down for decommissioning (1997)
La Hague R7 and T7 Vitrification Facilities	La Hague, France	Induction melting	1989 (R7 facility) 1992 (T7 facility)	The La Hague facilities are operational and are used to support spent nuclear fuel reprocessing.
Waste Vitrification Plant (WVP)	Sellafield, U.K.	Induction melting	1991	WVP is operational and is used to support spent nuclear fuel reprocessing.
Pamela Vitrification Facility	Mol, Belgium	Joule heating	1985	Facility shut down for decommissioning (1991)
West Valley Demonstration Project (WVDP)	West Valley, New York, U.S.A.	Joule heating	1996	Facility shut down for decommissioning (2002) [Meess 2011]
Defense Waste Processing Facility (DWPF)	Savannah River Site, Aiken, South Carolina, U.S.A.	Joule heating	1996	Operating
Tokai Vitrification Facility (TVF)	Tokai, Japan	Joule heating	1995	Shut down
Mayak Vitrification Plant	Mayak, S. Ural, Russia	Joule heating	1987 (I melter) 1991 (II melter)	Operating
Waste Immobilization Plant (WIP)	Trombay, India	Pot glass induction melting	1986	Operating

*See Section 3.1.5.2 for the vitrification technology description.

3. OPERATIONS AND SYSTEMS

In the U.S., the majority of spent fuel is a product of operating light-water reactor plants. Presently, the enrichment of fresh fuel is up to approximately 5%, and the typical burnup of spent fuel is about 60 gigawatt-days per metric ton of uranium. For fuel reprocessing, spent fuel is cooled for at least 5 years, although the majority of fuel in the waste stream is significantly older. Accounting for the range of fuel types and ages in the design and operating procedures for a fuel reprocessing facility is essential.

There are four main separation processes that are either in use or proposed for future use. Briefly, these are the following:

1. UREX Process – This process is similar to the PUREX process except that the chemistry is adjusted so that uranium and technetium are extracted in two product streams while product waste stream contains transuranics and fission products.

2. PUREX Process –This process uses nitric acid for dissolution and tributyl phosphate (TBP) as an organic solvent [Pace 2006]. This process is commonly used to extract plutonium for use in weapons or mixed oxide fuel for reactors and forms the base separation process for fuel reprocessing in the world today.

3. Co-Extraction Process – This process was developed by CEA (Commission of Atomic Energy in France) and AREVA, and is a modified version of the PUREX process. It is divided into two main separation phases: a) extraction cycles for separating and purifying U-Pu mixtures and b) forming a U-Pu nitrate solution to convert U and Pu into (U, Pu)O_2 [NUREG-1909]. A detailed AREVA process description is provided in [Bader 2011].

4. NUEX Process – This is also a modified PUREX separation process producing four outputs: (a) recycled uranium, (b) mixed plutonium, neptunium, and uranium to enhance proliferation resistance, (c) mixed americium and curium, and (d) a waste stream for vitrification containing the remainder of the spent fuel isotopes [Energy Solutions 2008].

Currently, as indicated in the previous section, there is no operating spent fuel reprocessing facility in the U.S. Section 3.1 describes a reprocessing facility based on the co-extraction process as an example of a facility that might be built in the U.S. Section 3.1 also describes the typical operations of a co-extraction facility. Section 3.2 describes the typical physical systems that can be found in such a facility.

3.1 Operations

In most spent fuel reprocessing facilities, the following operations are typical (illustrated in Figure 3-1):

1. Receiving and shipping
2. Storage (spent fuel pool and fuel container processing)
3. Crushing/shearing/chopping (processing of spent fuel)
4. Chemical processing
   a. Extraction
   b. Separation

c. Purification

d. Solvent recovery

e. Enrichment (optional)

5. Waste management, which includes waste storage and vitrification
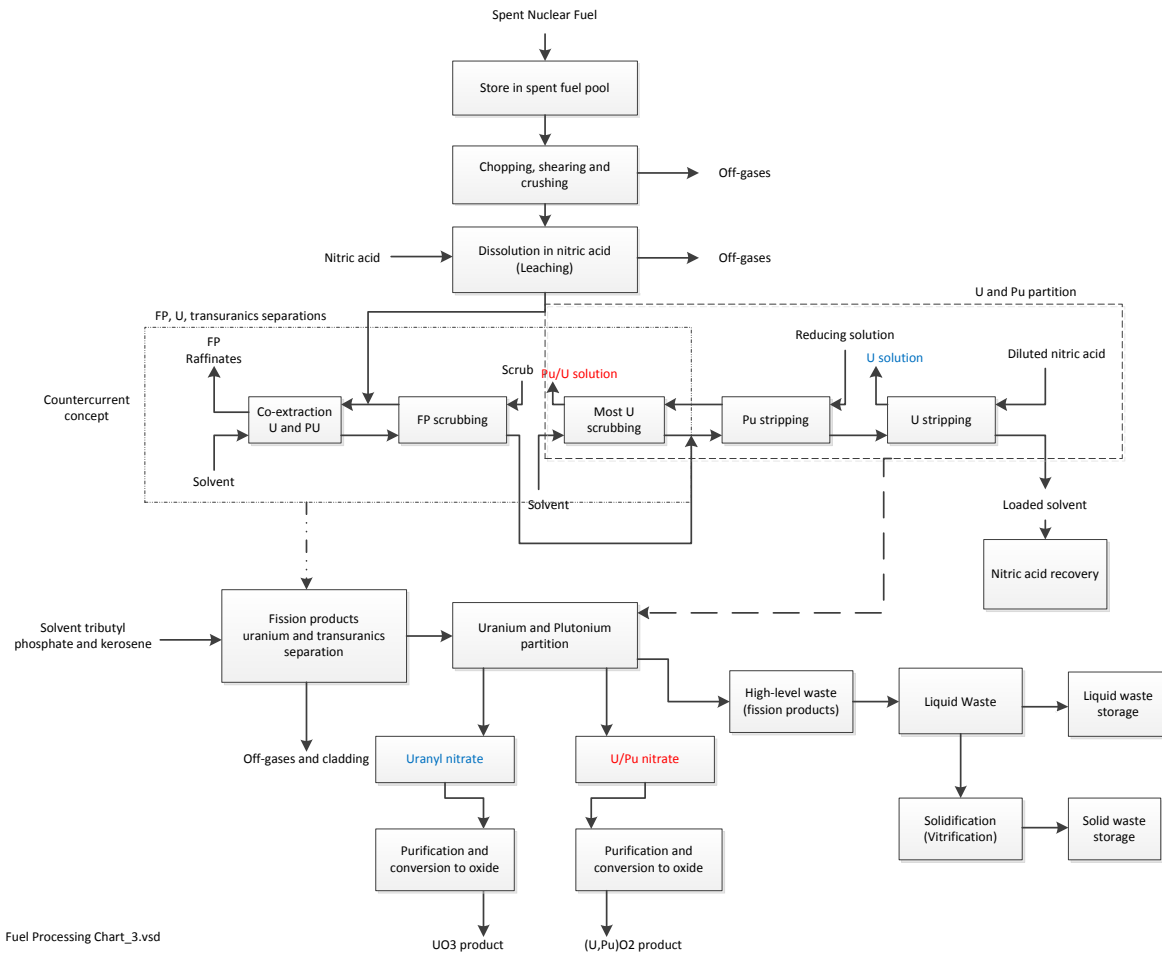
Figure 3-1   Typical operations at spent fuel reprocessing facilities that uses a co-extraction process with countercurrent concept[1]

The following subsections provide a brief discussion of the processing facilities in which the operations described above are performed.

3.1.1   Shipping and Receiving Operations

Shipping and receiving operations use certified shipping packages. Once spent fuel arrives at the facility, it may be moved to a spent fuel pool where the spent fuel is stored until reprocessing begins.

---

[1] Note that the countercurrent flow diagram of the extraction phase was adapted from [Todd 2008], but it has been enhanced for the co-extraction process. A detailed description of each operation is presented in subsequent sections.

3.1.2　　　　Storage

Fuel reprocessing facilities have an area designed to store incoming spent fuel ready for reprocessing. This can be in dry storage or in a spent fuel pool. Since the spent fuel is allowed to cool for at least five years before being accepted for reprocessing, the decay heat of the spent fuel stored in the spent fuel pool at a fuel reprocessing facility is typically lower than that stored in the spent fuel pool at a nuclear power plant. Potential events that can occur in a shipping and receiving facility are mostly from dropping of shipping packages, fuel assemblies, or fuel rods.

3.1.3　　　　Crushing, Shearing, and Chopping

Spent fuel assemblies are chopped, crushed, or sheared to increase the surface area for interaction between solvent and fuel. During this operation, fission product gases may be released and thus adequate ventilation and filtration systems must exist to capture these gases. Accidents related to oxidation of cladding and fuel can occur, especially when the interactions with air occur at elevated temperatures. Because of the high radiation level associated with spent fuel, this operation is often performed behind a shielded wall or in a hot cell where the ventilation and atmosphere are controlled. This portion of the facility has a potential for fires involving pyrophoric metals. A loss of ventilation/atmosphere control could also lead to a radioactive release.

3.1.4　　　　Extraction and Separation

There are several separation processes employed in spent fuel reprocessing plants. All but the electrochemical process involves dissolution of spent fuel and cladding materials as well as extraction of certain constituents for further processing. Below is a list of chemical processes that have been developed for use since World War II:

1.　　　　Bismuth Phosphate Process – This process was developed during World War II with the specific purpose of recovering plutonium (uranium is not recovered). Six steps are used in this process: (1) fuel dissolution in nitric acid, (2) adjustment of plutonium to the tetravalent state Pu(IV) with ferrous iron or nitrite, (3) addition of bismuth and phosphoric acid to separate Pu(IV) and $BiPO_4$ from uranium, aluminum, and most fission products, (4) re-dissolution of the Pu in highly concentrated nitric acid, (5) incorporation of dichromate to oxidize plutonium to the hexavalent state, and (6) precipitation of $BiPO_4$ to obtain Pu(VI) [Campbell 1990]. Steps (2) to (6) can be repeated until the desired purity is achieved. This process was used at Hanford starting in late 1944 [Gephart 2003].

2.　　　　Ion Exchange Process – This process was developed about the same time as the Bismuth Phosphate Process. This process uses sulfonic acid and organic cation exchange resins. A distinct disadvantage of this process is that it is difficult to obtain a highly pure Pu product [Campbell 1990].

3.　　　　REDOX Process – This process uses nitric acid and aluminum nitrate for dissolution and hexone (methyl isobutyl ketone) as an organic solvent [Pace 2006]. It is a continuous process to recover both U and Pu with high yield and high purity. The process was developed at Argonne National Laboratories, and tested in a pilot plant at Oak Ridge in 1948-49. A REDOX plant was built at Hanford in 1951. Hexone is highly flammable and volatile [Todd 2008].   Hexone is also unstable in high concentrations of nitric acid and a

large amount of neutral salt, such as aluminum nitrate, is required to maintain chemical stability. Because of the waste management issue related to large volumes of waste, a process with emphasis on recovered and recycled waste is preferred [Campbell 1990].

4.   BUTEX Process – This process uses dibutyl carbitol as a solvent and nitric acid as a salting agent. This method eliminates the need for aluminum nitrate used in the REDOX process.

5.   PUREX Process –This process uses nitric acid for dissolution and tributyl phosphate (TBP) as an organic solvent [Pace 2006]. This forms the base separation process for fuel reprocessing in the world today. This process was used at Hanford in the PUREX plant starting in the early 1950s [Gephart 2003].

6.   Co-Extraction Process – This process was developed by CEA and AREVA , and is divided into three main phases: a) extraction cycles for separating and purifying U-Pu mixtures, b) forming a U-Pu nitrate solution to convert U and Pu into $(U, Pu)O_2$, and c) a powder metallurgy process to produce the fresh mixed oxide fuel [NUREG-1909]. A detailed AREVA process description is provided in [Bader 2011].

7.   NUEX Process – This is a separation process producing four outputs: (a) recycled uranium, (b) mixed plutonium, neptunium, and uranium to enhance proliferation resistance, (c) mixed americium and curium, and (d) a waste stream for vitrification containing the remainder of the spent fuel isotopes [Energy Solutions 2008]. The NUEX process contains three primary separation processes: (a) Primary separation using TBP dissolved in odorless kerosene, similar to the primary process used in THORP; (b) TRUEX (Transuranic Extraction); and (c) TALSPEAK (Trivalent Actinide-Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes). Similar descriptions are given elsewhere [NUREG-1909].

8.   FLUOREX [Ogata 2003] – A hybrid system of using the fluoride volatility process to recover most of uranium prior to the conventional PUREX process, where the residual uranium and plutonium are purified (see PUREX process above). This process is being employed at the Rokkasho Reprocessing Plant.

9.   Electrochemical [NUREG-1909] – This process requires that any oxides in the spent fuel be reduced to metallic form – either chemically by molten lithium or by electrolytic reduction in molten lithium chloride. The metal from oxide reduction or metallic spent fuel including cladding materials becomes the anode in an electro-refiner. This refiner contains a molten electrolyte salt (a mixture of LiCl and KCl) atop of cadmium metal. For an operating condition of 12 hours, the products from the electro-refiner contain: an anode with elements that are stable as metals (e.g., zirconium, technetium, iron, and molybdenum), a cathode containing most of the uranium as metal, and a cathode containing TRU elements as metal, and rare earth fission products. The molten salt contains most of the fission products as chlorides.

3.1.5   Waste Management

There are a number of waste streams produced by fuel reprocessing. High-level waste may be stored onsite or placed in a certified shipping container to be shipped to other sites to be

processed. If the facility is capable of enrichment, the processed uranium and plutonium may be further enriched. If not, these materials are placed in certified Special Nuclear Material (SNM) containers to be shipped elsewhere for further enrichment. Other residual chemicals may also be shipped to other facilities.

After the spent fuel has been processed, the product, waste, and recycle streams may require storage prior to being shipped, subject to further reprocessing, or re-use in operation processes. Recycled acid must be contained in a robust and noncorrosive container. The end products, uranium oxide and plutonium oxide/mixed oxide, can be stored in a certified shipping container to be shipped out to fuel fabrication or enrichment facilities. The high-level waste can be stored on-site in solidified form or for further reprocessing of the waste. Storage-related accidents are possible, especially for high-level liquid waste, in which radiolysis of aqueous solutions may result in high concentrations of hydrogen. Inadequate ventilation can lead to explosions.

As a byproduct, the waste stream from spent fuel reprocessing contains most of the non-usable fission products and impurities that may not be suitable for further reprocessing. Some streams may be separated into low-level, intermediate, and highly radioactive liquid waste. Liquid waste is stored in tanks, while any processed waste in solid form can be vitrified. Similarly, some types of liquid waste may be vitrified once dried. The vitrification process combines solid waste with glass frit, which is placed into a melter for heating until a glass is formed. The vitrification process has been used extensively for immobilizing waste for long-term disposal options.

The potential accidents associated with the waste operation include:

1.   Storing liquid waste – potential H_2 explosion (due to radiolysis of water) and leaks in tanks, valves, and pipes

2.   Drying and solidification of the liquid waste – external heating that could cause runaway chemical reactions and possibly gas explosions

3.   Loading of the waste into containers – spills and potential fires

4.   Loss of cooling of high-level waste (HLW) storage tank

Note that human errors/machine malfunctions typically cause the accident conditions during loading of waste into the containers.

3.1.5.1   Liquid Waste Storage

Onsite liquid waste storage is often needed as part of the fuel reprocessing operation. Several types of liquid waste can be handled that have low, intermediate, and high levels of radioactivity. High-level liquid waste requires shielding and additional equipment and monitoring systems to ensure that hydrogen buildup due to radiolysis is controlled. Portions of these liquid wastes may be further processed into solid forms, as discussed in the next section.

3.1.5.2   Drying and Solidification (Vitrification)

In order to be solidified, liquid waste must undergo drying to remove solvent. This drying generally requires heating. First, a facility may use a concentrator to reduce the amount of solvent and to increase the concentration of the waste. Then, an evaporator may be used to further reduce the solvent content. Finally, a denitrator (or calciner) is used to completely

remove solvent. Once dried, the waste is subjected to a solidification process. The solidification process is often referred to as vitrification, which immobilizes the waste material in glass form. There are other non-vitrification solidification technologies that use sintering or a molten metal process [Jain 1998b]. Other solidification processes, such as for uranium and plutonium, are discussed in the description of the extraction cycle and in the descriptions of the Barnwell spent fuel reprocessing plant in Appendix A, and a recently proposed plant from AREVA [Bader 2011]. This section only describes the vitrification technology for creating solid waste forms.

Vitrification processes demonstrated or used by the Department of Energy (DOE) include: the joule-heated melter process, plasma arc centrifuge treatment (PACT) system, plasma hearth process (PHP), American Society of Mechanical Engineers (ASME)/United Status Bureau of Mines (USBM) arc melter furnace, and in-situ vitrification (see Table 3-1 for a description of each of these processes). Among these vitrification processes, only joule-heated melters have been used on a production scale at DOE.

Table 3-1 Solidification Technology [DOE 1999]

Technology*	Description	Comments
Joule-heated melter	This technology utilizes the melter furnace that incorporates a process of heating the glass by conducting electric current through the glass matrix. Both the waste concentrate and the glass frit (glass former) are directly introduced into the melting chamber. (See Figure 3-2 for the conceptual design of this technology.)	This technology is well established and mature for the INCONEL melter used in the U.S., Japanese and Belgian plants over 20 years [Jain 1998b].
Induction melting [Jain 1998a]	The French AVM process (Atelier de Vitrification Marcoule) technology utilizes a 2-stage process that consists of a combination of rotary kiln (a calciner) and an induction-heated metal glass-melting crucible [Baehr 1989]. (See Figure 3-3).	This technology is used at both United Kingdom and French vitrification facilities.
Plasma arc centrifuge treatment (PACT)	This technology utilizes the heat transferred from a plasma arc torch to create a molten bath. In the process, the solids are melted into the bath (~1760 °C) while organics are evaporated and/or pyrolized and partially oxidized. The off-gas leaving the primary chamber enters the secondary combustion chamber (SCC), where a complete oxidation takes place. (See Figure 3-4 for the schematic).	This technology has been demonstrated under the Environmental Protection Agency superfund innovative technology evaluation.
Plasma hearth process (PHP)	This technology is intended to stabilize mixed wastes. It is a high-temperature thermal treatment process using a plasma direct-current arc torch in a stationary, refractory-lined melter. The plasma arc destroys organics and stabilizes residuals in a vitrified slag. The drummed waste is fed to the plasma chamber. Similar to PACT, the off-gas is transported to the cyclonic combustion chamber, where a natural gas burner is used to completely burn organics to yield CO_2 and water. (See Figure 3-5 for the prototype design.)	This technology was developed for the DOE mixed waste treatment.
Arc melter furnace	This technology utilizes a plasma arc furnace to pyrolyze combustible organic material and melt residual inorganic materials with an electric arc. This furnace contains three carbon electrodes, a continuous feed system, and an off-gas treatment system with slag- and metal-trapping capabilities. (See Figure 3-6 for the design).	This technology was used for the Phase 1 testing of Hanford low-level waste [Jain 1998b].

Technology*	Description	Comments
In-situ vitrification ISV)	This technology uses electrical power to melt in-situ contaminated earthen media such as soil, sediment, and mine tailings. The ISV process is initiated by formation of a pool of molten soil at the surface of a treatment zone between four electrodes.	This technology has been evaluated and implemented within the DOE and other federal agencies, and aboard [Jain 1998b]. It has largely been rejected as a viable approach.

* There are other technologies that are not included in this table. The reader is encouraged to consult [Jain 1998a] and [DOE 1999].
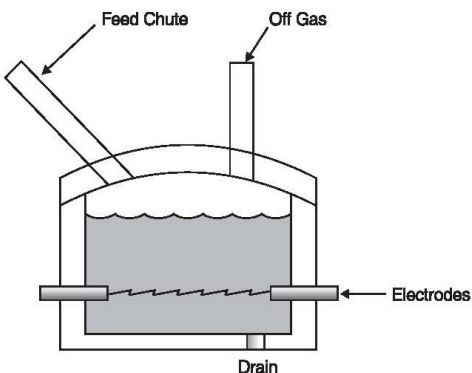
Figure 3-2   Joule-heated melter concept [DOE 1999]
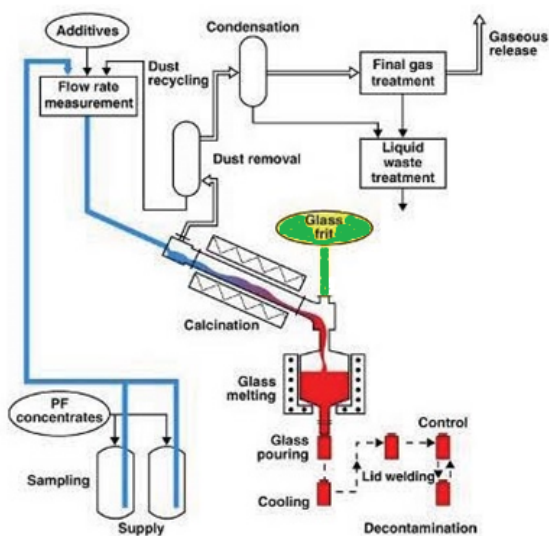
Figure 3-3   Induction melting used at the French AVM process [Marsden 2010]
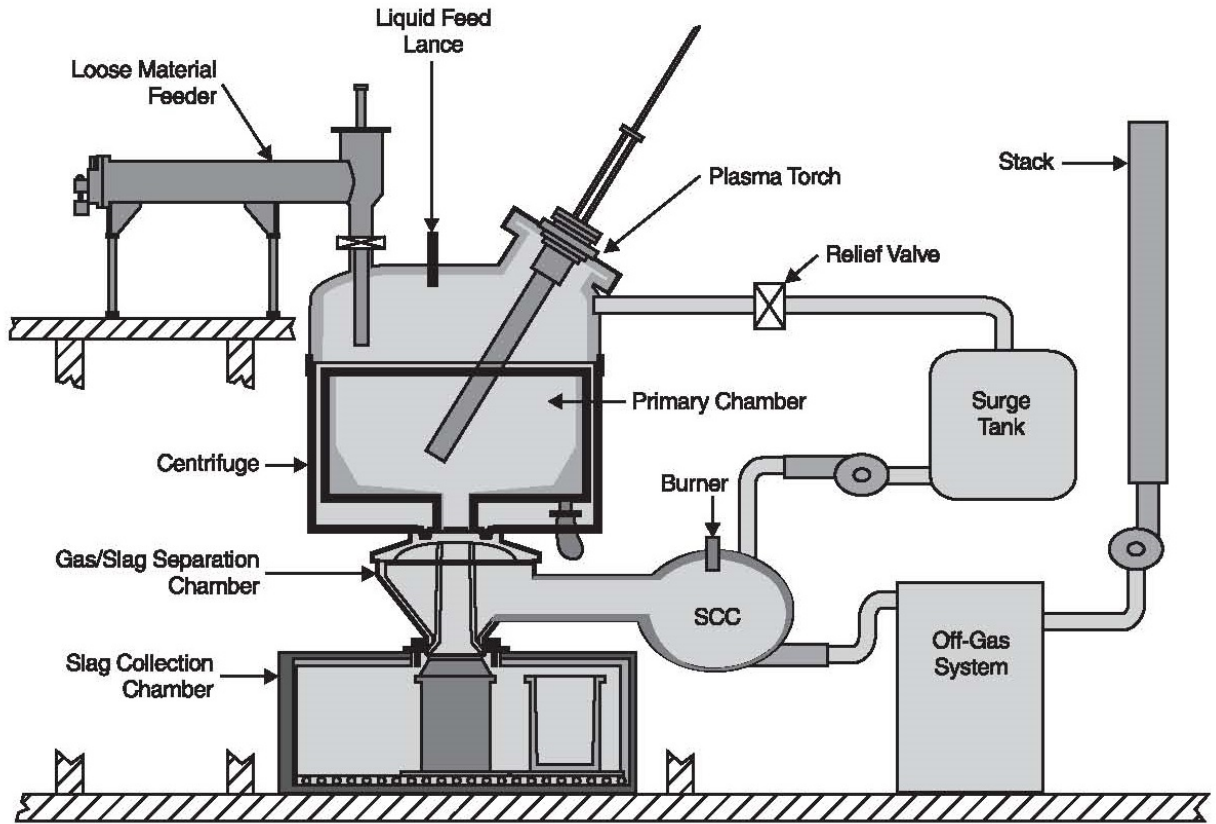
Figure 3-4 A typical component list of a PACT melter [DOE 1999]
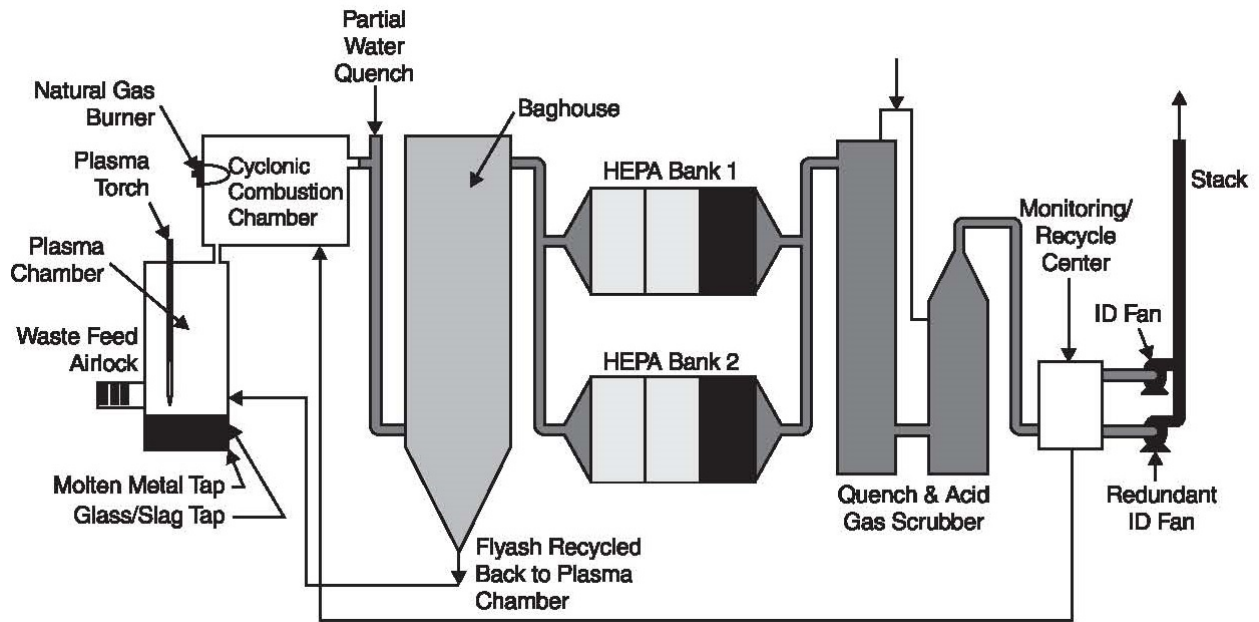
Figure 3-5 PHP prototype design [DOE 1999]
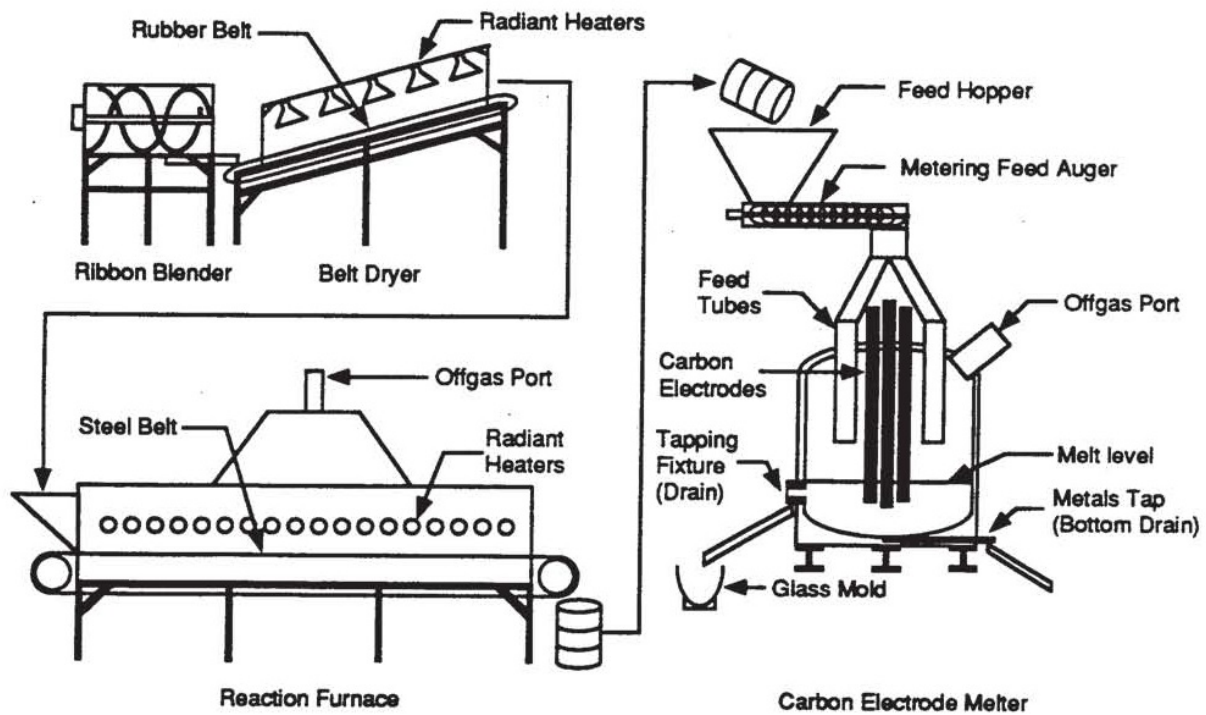
Figure 3-6   Feed preparation and melter schematic for the arc melter furnace design [Jain 1998b]

The following processes are included within the vitrification operation:

1.   Waste pretreatment and feed preparation
2.   Vitrification
3.   Off-gas treatment system

Many common problems facing the development of a reliable and safe melter technology for the vitrification of waste include [DOE 1999]:

- Inadequate design considerations for maintenance and surveillance

- Lack of a comprehensive failure modes and effect analysis

- Lack of a systems engineering analysis on design and construction

- Leakage through penetrations (for instrumentation, drains, etc.) of the melter shell(s) and liner, which are typical weak spots

- Clogging of the melt discharge orifice

- Low reliability of critical support systems, including cranes, water cooling systems, remote mechanical equipment, and maintenance support systems.

- Corrosiveness of the waste materials, especially where electrodes are concerned in joule-heated melters

- Incompatibility of the refractory lining materials for the melter with the waste/glass-forming materials

3.2   Systems

The following physical systems are included in a typical spent fuel reprocessing facility:

1.   Confinement – This system provides the mitigation and/or prevention of an environmental release of radioactive elements and chemical products. This system also has a significant impact on the safety of the plant, particularly the dose to onsite employees as well as potential dose offsite.

2.   Ventilation and filtration – This system is designed to reduce radioactive and chemical contamination of various atmospheres within the facility through the use of filters. A negative pressure differential maintained by the ventilation system reduces offsite leakage through environmental pathways. Ultimately, the ventilation system processes the air released to the environment to maintain acceptable radiation and chemical releases. This system is also important to worker safety.

3.   Fire suppression and detection – This system is used to mitigate the spread and duration of a fire and to reduce the suspension of radioactive isotopes and harmful chemical species.

4.   Miscellaneous support systems – These systems can include electrical power, including backup power for the plant, chemical systems supporting various plant operations, and water resources that are used in the extraction and cleaning processes.

All of these systems are engineered safety systems and provide potential mitigation of an accident inside the reprocessing facility. Each is described in more detail in the subsequent sections. Power backup is important since many of the ventilation/filtration systems and fire sprinklers may rely on this system if offsite power is lost.

The NRC has published a number of guidance documents for the systems in fuel reprocessing plants (see Table 3-2).

Table 3-2   Selected Nuclear Regulatory Commission guidance documents for fuel reprocessing[2]

Regulatory Guide No.	Title [Reference]	Safety Functions or Pertinent Information
3.3	Quality Assurance Program Requirements for Fuel Reprocessing Plants and for Plutonium Processing and Fuel Fabrication Plants	• 10 CFR Part 50 Appendix B establishes quality assurance requirements for nuclear power plants and fuel reprocessing plants.

3.6	Content of Technical Specifications for Fuel Reprocessing Plants	• Technical specifications should include both technical and administrative matters.
		• The use of safety limits and limiting control settings for establishing technical specifications.
		• In terms of administrative matters, four principal functions should be performed:
		  o Operation of plant equipment
		  o Maintenance of equipment
		  o Record keeping
		  o Audits, reviews, and evaluation of operations

3.17	Earthquake Instrumentation for Fuel Reprocessing Plants	• Identification of the locations and requirement of earthquake detection instruments and recorders for the structures, systems and components important to safety.
3.18	Confinement Barriers and Systems for Fuel Reprocessing Plants	• Confinement systems minimize the radioactive materials to restricted areas according to the limits specified in §20.103 of 10 CFR Part 20. • These systems should minimize the radioactive materials to unrestricted areas according to the annual limit prescribed by § 20.106(a) of 10 CFR Part 20.
3.19	Reporting of Operating Information for Fuel Reprocessing plants	• Establishment of the reporting requirement, including types and time intervals of the documents submittal.
3.20	Process Off -Gas Systems for Fuel Reprocessing Plants	• All parts of the process off-gas system should be designed to limit the radioactive and noxious materials release • The design must withstand postulated accident conditions • Backup systems should exist to prevent any single failure
3.21	Quality Assurance Requirements for Protective Coatings Applied to Fuel Reprocessing and to Plutonium Processing and Fuel fabrication Plants	• This guide provides quality assurance requirements for protective coatings over ferritic steels, aluminum, stainless steel, zinc-coated (galvanized) steel, concrete or masonry surfaces of the plants.
3.22	Periodic Testing of Fuel Reprocessing Plant Protection System Actuation Functions	• The protection system that is important to safety should be designed to permit periodic testing to extend to and include the actuation devices and actuated equipment.

[2] NRC guidance documents may need to be updated to support licensing of a modern reprocessing facility.

Table 3-2   Selected Nuclear Regulatory Commission guidance documents for fuel reprocessing[2]

Regulatory Guide No.	Title [Reference]	Safety Functions or Pertinent Information
3.26	Standard Format and Content of Safety Analysis Reports for Fuel Reprocessing Plants	• This guidance provides the format and content to document a safety analysis report for fuel reprocessing plant.
3.27	Nondestructive Examination of Welds in the Liners of Concrete Barriers in Fuel Reprocessing Plants	• This guide provides the requirement to examine (nondestructively) the welds for the liners of concrete structures. The liners are important to mitigate any spills inside the facility.
3.28	Welder Qualification for Welding in Areas of Limited Accessibility in Fuel Reprocessing Plants and in Plutonium Processing and Fuel Fabrication Plants	• This document describes the welder qualifications for weld fabrication and repair of the safety related structures within the fuel reprocessing facility.
3.29	Preheat and Interpass Temperature Control for the Welding of Low-Alloy steel for Use in Fuel Reprocessing Plants and in Plutonium Processing and Fuel Fabrication Plants	• This document describes a method that meets the temperature-control requirements for the welding of low-alloy steel components for fuel reprocessing, plutonium processing and fuel fabrication plants.
3.30	Selection, Application, and Inspection of Protective Coatings (Paints) for Fuel Reprocessing Plants	• This guide provides a standard for evaluating and selecting protective coatings (paints) for nuclear plants. • This guide also provides guidance for preparing the surfaces to be coated and for applying and inspecting the coatings under both shop and field conditions.
3.31	Emergency Water Supply Systems for Fuel Reprocessing Plants [NRC 1975b]	• This guide discusses provision of water to meet the heat rejection requirements for the fuel storage pool, waste storage tanks, and certain process vessels in abnormal conditions. • It also discusses provision of water for the fire protection system in case the normal supply is disrupted, and • It requires that water must be provided for any safety related equipment or systems in accident conditions.
3.32	General Design Guide for Ventilation Systems for Fuel Reprocessing Plants	• This guide provides the requirements for a well-designed ventilation system for fuel reprocessing plants.
3.37	Guidance for Avoiding Intergranular Corrosion and Stress Corrosion in Austenitic Stainless Steel Components of Fuel Reprocessing Plants	• This guide documents the guidance and steps that can avoid inter-granular and stress corrosion in austenitic stainless steel components used in fuel reprocessing.

Table 3-2   Selected Nuclear Regulatory Commission guidance documents for fuel reprocessing[2]

Regulatory Guide No.	Title [Reference]	Safety Functions or Pertinent Information
3.38	General Fire Protection Guide for Fuel Reprocessing Plants	• This guide documents a requirement to establish a fire protection program within the facility to minimize and mitigate the fire accidents in the facility. It also provides steps to mitigate fire situation with fissile materials presented and critical control equipment presented.
3.40	Design Basis Floods for Fuel Reprocessing Plants and for Plutonium Processing and Fuel Fabrication Plants	• This guide describes methods of determining the design basis floods for nuclear facilities (fuel reprocessing, plutonium processing and fuel fabrication plants).

3.2.1   Confinement

One of the most important safety features in a spent fuel reprocessing plant is the confinement system. The building enclosing the extraction or storage operations is designed to minimize radiation exposure to the workers and potential airborne release to the public and environment. Typically, the confinement requirements are based on radiation levels for standard operations. Table 3-3 shows the working environment enclosure requirements as a function of the radiation level, mainly for inhalation and direct exposure.

Table 3-3 Confinement needs for handling radioactive materials [Cadwallader 2005]

Work Environment	Radioactivity Level (ALI*)	Radioactive Quantity (Curies)
Laboratory bench top	< 0.1	$< 10^{-9}$
Laboratory fume hood	0.1 to 10	10^{-9} to 10^{-6}
Unshielded glove box enclosure	10 to 1,000**	10^{-6} to 10^{-3}
Shielded glove box enclosure	10 to 1×10^{6}	10^{-3} to 10^{0}
Hot cell	> 1×10^{6}	10^{0} to 10^{6}

 * ALI stands for annual limits on intake.

 ** The glove box may protect radiation level > 1,000 ALI if the emitted radiation is α or β particles rather than ɣ or neutron. If the radiation is penetrating, then a shielded glove box may need to be used at a small ALI value.

For direct radiation exposure, the use of physical barriers such as concrete walls, are used to reduce worker exposure from extraction operations and high-level waste storage cells.

To mitigate the release of airborne material during an accident, multiple-tiered confinement buildings are generally used. Figure 3-7 shows a 3-tiered confinement system. As shown in this figure, the primary confinement/barrier (Zone I) has either a storage container, process vessel, glove-box, or processing equipment that contains the radioactive and/or toxic materials. The primary confinement may be contaminated and is treated as such. To contain the contamination if Zone I is breached, the primary confinement/barrier is contained within a secondary confinement/barrier (Zone II), which is physically an operation room or cell. Depending on the contamination/acidity level expected in this secondary confinement/barrier, the floor, walls, and ceiling may be designed to protect building structures from chemical

interactions caused by leaks or spills. This is usually done by using a nonreactive liner such as stainless steel. In most extraction operations, the processes are performed inside hot cells that have walls thick enough to shield workers from direct exposure to radiation from the spent fuel.
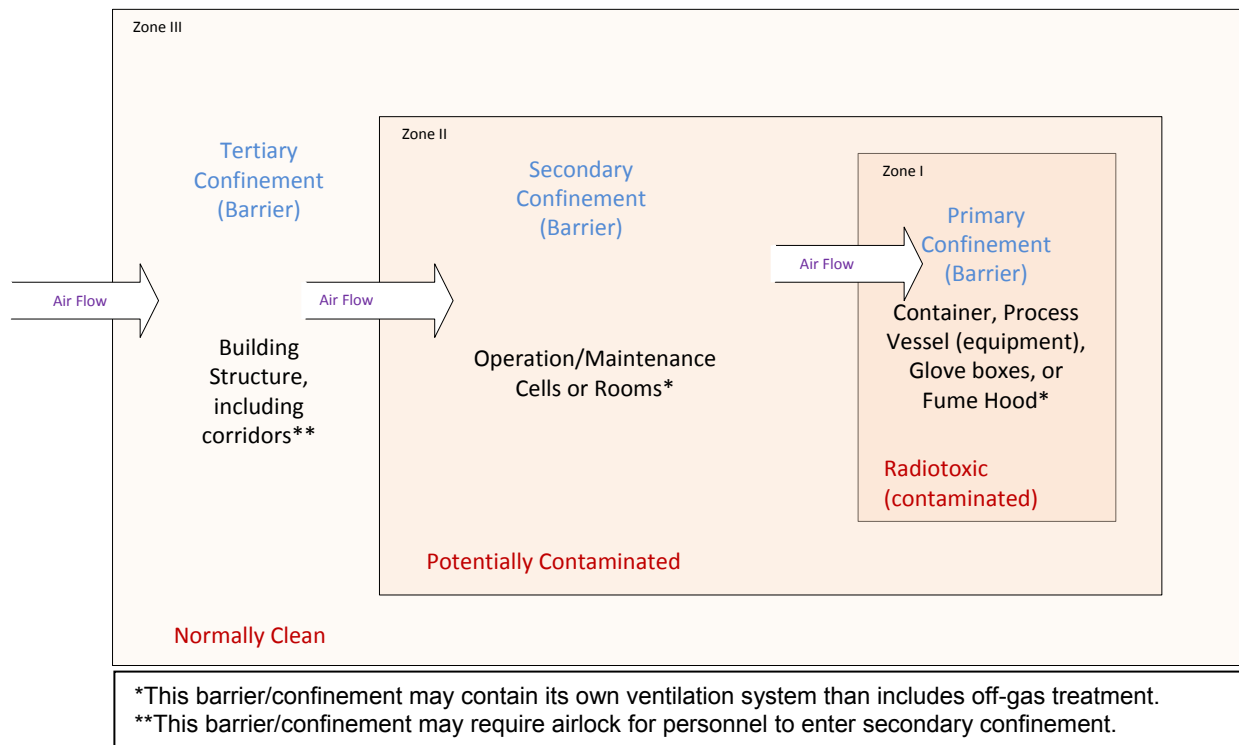
Figure 3-7   Three-tiered confinement zones used in radiological plant

In most instances, the secondary confinements/barriers reside within a third building structure that acts as a tertiary confinement/barrier (Zone III). For example, the Barnwell Nuclear Fuel Plant was designed with 2 to 4 barriers. In this plant, all chemical extraction operations are done inside hot cells for which the floors and walls are lined with stainless steel and contain a sump for spill collection. The hot cells are contained in the main process building. All hot cell walls are thick enough to minimize worker exposure outside the cells, such as in operating corridors.

Unlike nuclear power plants for which the containment is designed to withstand internal or external challenges such as high internal pressure and temperature and airplane crashes, many spent fuel reprocessing plants contain only confinement systems that may not be capable of withstanding any high energy accidents, such as an explosion inside or outside the facility.

3.2.2   Ventilation

The ventilation systems provide pretreated airflow to the processing areas, and maintains pressure differentials between zones as required to confine radioactivity to designated locations in the plant (see Figure 3-7). The ventilation system is designed to confine and channel any airborne release to treatment systems such as filters and off-gas treatment systems to capture volatile fission products or other toxic gases before they can exit via the exhaust stack. For example, the Engineering Alternative for Extraction study [Bader 2011] shows that the process

vessel ventilation system is used to treat any off-gas from the operations to ensure that no significant amount of either gaseous (or volatile) fission products or toxic gases are released. Cooling and recirculation for the process cells are also provided by the ventilation systems.

Maintaining proper pressure differentials between the processing cells and the corridors or between the glove box and the operating cell is essential to keeping contamination within its intended confinement. Table 3-4 and Table 3-5 show the permissible quantity of materials within each confinement zone. Similar permissible levels of radioactivity and toxicity of materials are expected for a commercial reprocessing facility per NRC regulations [NRC 1979].

Table 3-4 Zoning of facilities based on radioactivity of materials handled [DOE 2003]

Quantity of Material Permitted in Zone at Any One Time*			
Radiotoxicity of Isotope	*Primary Confinement*	*Secondary Confinement*	*Tertiary Confinement*
Very High	> 10 mCi	0.1 µCi – 10 mCi	0-0.1 µCi
High	> 100 mCi	1.0 µCi – 100 mCi	0-1.0 µCi
Moderate	> 1 Ci	10 µCi – 1 Ci	0-10 µCi
Negligible	> 10 Ci	100 µCi – 10 Ci	0-100 µCi

*There are practical upper limits to the material quantity in any particular zone, based on the material type and design of the confinement system. For example, criticality safety concerns may restrict the amount of fissile material that can be handled at one time, fire protection concerns may limit the amount of pyrophoric materials, and shielding considerations may limit the amount of materials when penetrating radiation is emitted. The above criteria are based on the potential for the activity to generate airborne radioactive materials.

Table 3-5 Zoning of facilities based on chemical contamination levels [DOE 2003]

Anticipated Contamination Levels			
Type of Contamination	*Primary Confinement*	*Secondary Confinement*	*Tertiary Confinement*
Airborne [a]	> 100 × DAC	1 × DAC – 100 × DAC	< 1 × DAC
Removable surface [b]	>> RSCV [c]	> RSCV [c]	< RSCV

[a] The DAC is the derived airborne concentration value listed in 10 CFR 835, Appendix A for the type and chemical form of the material being handled.
[b] The RSCV is the removable surface contamination value listed in 10 CFR 835, Appendix D, for the type of the material being handled.
[c] RSCV level do not always directly lead to an increasing level of airborne contamination. The level of airborne contamination strongly depends on the potential for the particular activity to re-suspend the deposited particles into the atmosphere.

To capture any airborne release in the ventilation system, the ventilation system must contain the proper equipment such as high efficiency particulate air (HEPA) filters, charcoal filters, and sand filters). There are generally several ventilation systems supporting each confinement zone in the plant.

Off-Gas System - This system (Zone I) is used in conjunction with process operations in which significant amounts of airborne and gaseous radioactive and toxic materials may be present (see Figure 3-7 for Zone I definition). In the proposed AREVA plant [Bader 2011], the off-gas system is used to remove noble and volatile fission products such as xenon, krypton, iodine, ruthenium, tritium, and carbon-14. Additionally, NO_x must also be removed in the off-gas system before the effluent is released through the exhaust stack. Removal and trapping processes used in off- gas systems are also found in the proposed AREVA plant [Bader 2011].

Depending on the removal system used, the decontamination factor may be reduced when there are impurities in the gas stream. For example, an off-gas system used in the extraction operation may contain silver mordenite, an iodine sorbent to capture airborne iodine. However, experiments have shown that the decontamination factor of the iodine by this sorbent may be reduced in the presence of the airborne TBP vapor [Parker 1980].

Process Room System – This system is used to provide ventilation in the process cells or rooms where the spent fuel reprocessing operation resides. In most cases, the operations take place inside a hot cell, where the operator behind external walls uses manipulators to conduct operations. Within the process room, the atmospheric pressure is higher than that of the off-gas system for the processing equipment or process vessel, so that contamination in the process room is controlled. In the event of a chemical fire, the process room atmosphere may be inerted.

Filter System – This system is a safety system that is used to capture airborne materials including an abnormal release of airborne radionuclides. The airborne materials may be in the form of vapors or aerosol particles. The filter systems can be located at the inlet and/or exhaust portions of the ventilation systems. Inlet filters are used to trap aerosol particles that enter the facility. Exhaust filters also trap aerosol particles that are generated during normal operations such as where crushing or chopping of the spent fuel assemblies takes place. During abnormal situations, filters are designed to be able to decontaminate 99.97% of 0.3 μm airborne materials and are designed to survive the accident.

Zone II System – This system may include the Process Room System previously described, and is used to provide a secondary confinement zone so that airborne release from Zone I to Zone II is minimized (see Figure 3-7).

Zone III System – This system is the outermost ventilation system that keeps the recirculation of the air clean for areas occupied by workers. This area includes maintenance rooms and corridors.

Construction materials for the ventilation ducting are also important. These ducts may need to be able to withstand acidic environments, especially the ducts connected to the rooms or chambers with extraction equipment.

3.2.3   Fire

Unlike a nuclear power plant, where spray systems in the containment are used to reduce the pressure from an energetic accident and wash down fission products, fire suppression systems are primarily designed to mitigate fires. However, these sprays can also reduce airborne vapors and aerosol particles. Fire detection systems do not perform the same function directly, but can facilitate human interdiction were a fire to occur.

Fire sprinkler systems are active engineered safety features in a spent fuel reprocessing facility. They require a water source, pumps, and electrical power in order to function. Because the amount of fissile materials processed in the fuel reprocessing facility is often large, this system must be designed to fight fire without inducing inadvertent criticality. The use of Halon fire suppression systems may be considered in areas where water could induce additional consequences.

Other than the process operations, sprinkler systems may be used in a storage building, pump houses, and in ventilation systems where filters are located.

3.2.4 Miscellaneous Support Systems

There are a number of miscellaneous support systems in a fuel reprocessing facility. These systems include:

- Electrical backup system – the function is to provide backup power in the case of loss of offsite power. It must be able to provide adequate power to activate engineered safety systems and cooling to processes that contain materials with high decay heat. Typical backup systems include standby diesel generators that can provide power following a loss of offsite power.

- Chemical supply system – the function is to provide the fresh chemicals and recycled chemicals to support extraction and solidification operations in the facility.

- Water system – the function is to provide water to support various operations in the facility, which can include:

  - Spent fuel pools
  - Steam to support concentrators, evaporators, denitrators, and calciners
  - Extraction processes
  - Fire-fighting

- Process control instrumentation – the function is to provide information to the plant operators to evaluate the conditions in various processes in the facility, which may include:

  - Pressure control
  - Temperature control
  - Concentration control
  - Volume control

4. ACCIDENTS/INCIDENTS – HISTORY AND DISCUSSION

This section describes potential accidents or incidents that could occur at spent fuel reprocessing facilities (see Table 4-1). Reprocessing facility accidents involve spills, fires, explosions, and inadvertent criticality events; incidents are more minor occurrences. As shown in Table 4-1, the potential incidents are identified for each specific operation at the facility. The consequence of each incident is categorized in this table. Because of the types of chemicals used in the separation processes, chemical reactions that may lead to fire and explosions are included as possible accidents. In addition, because of radiation emitted from the spent fuel, any radiolysis process such as those involving water, can create flammable gases such as hydrogen that can lead to a deflagration or detonation. Inadvertent criticality events are possible since fissile material from the spent fuel is extracted and concentrated during the recovery process. A failure of the physical boundary of the processing equipment can permit a release. If the contents are liquid, a spill accident is also possible. These major anticipated accidents are identified and described in more detail in Table 4-2.

Table 4-1 Potential incidents at a spent fuel reprocessing facility [DP-1558]

Process	Low Consequence	Intermediate Consequence	High Consequence
Fuel Receiving and Storage Operation	• Fuel damage in transit or upon arrival • Fuel cooling time too short • Loss of cooling • Surface contamination not removed from cask • Low water level • Rupture of fuel during storage	• Cask inadvertently vented • Cask dropped • Damage to fuel assembly outside cask • Damage to stored fuel • Loss of cooling capability	• Hydrogen explosion in fuel cleaning vessel • Criticality
Shearing Operation	• Fuel jammed or stranded in shear • Irregular length of hulls • Shear jammed	• Malfunction of inert gas system • Chopped fuel overheating • Pressurization of shear • Fuel element overheating	• Pyrophoric fire[1] • Release of volatile and particulate activity into room
Dissolving Operation	• Un-complexed fluoride in solvent • Pu-rich residue settling in dissolver, lines, and other process vessels • Dissolver seal failure • Inadequate cladding rinse • Transfer error of dissolver solution • Suck back (due to eructation and pressurization of dissolver) • Siphoning	• Contact of sheared fuel at a temperature about 300 ˚C above boiling point of dissolver solution • Pressurization of the dissolver • Dissolver leakage (due to corrosion gasket/valve failure) • Charge of inadequately cooled fuel • Malfunction of dissolver off-gas (VOG) iodine adsorbers • Excessive interaction of fuel external to dissolver (slightly reduction in criticality safety margin) • Excessive pressure in water wash compartment except during basket exchange.	• Precipitation of Pu polymer (potential criticality) • Zirconium fire[1] • Zirconium explosion (due to significant fines in the dissolver)[1] • Explosion in the iodine adsorber • Overconcentration in dissolver (potential criticality) • Self-concentration of dissolver solution (potential criticality) • Low H^+/Pu ratio in dissolver solution (potential criticality) • Inadequate poison in dissolver or leach solution (potential criticality)

Table 4-1 Potential incidents at a spent fuel reprocessing facility [DP-1558]

Process	Low Consequence	Intermediate Consequence	High Consequence
		• Dissolver pot coils not submerged during shutdown (high release of volatiles to room atmosphere)	• High fuel loss in leached fuel fragments • Failure of cladding monitor (potential criticality) • Absence of basket in dissolver • Explosion in dissolver vessel or hold tanks (H_2 explosion)
Solvent Extraction Operation	• Plutonium concentration in a solvent extraction contractor increases beyond normal values • Potential backup of radioactivity by air lines • Flow reduction or interruption of scrub streams to contactors (uranium) • Loss of organic flow to a contactor (uranium) • Low temperature in scrub stream to contactor (uranium) • Low temperature in contactor • Emulsion in solvent washer • High plutonium losses in waste streams from contactors (in PUREX process) • High plutonium concentration in contactor (in PUREX process) • Excess gamma activity in plutonium product	• Potential for abnormal accumulation of fissile material by plutonium reflux in contractor, particularly during high activity waste rework (decreased criticality safety margin) • Potential for excessive product loss to spent organic effluent (reduced criticality safety margin) • Accumulated solvent in rooms • Solvent extraction system contents removed by overflow (in sump)	• Solvent fire[2] • Potential for buildup of unsafe amounts of fissile material in organic storage tank (potential criticality) • Potentially unsafe feed concentration (potential criticality) • Potential for excessive plutonium loss to aqueous waste (potential criticality) • Incorrect material transfer of feed chemicals, product materials or incorrect materials (potential criticality, excessive radioactive material release)
Product Evaporation	• Leaks in tank containing concentrated uranium-plutonium solutions	• Transfer error in a product evaporator system • Coil or tube-bundle (in reboilers and condensers) failure • Overflow of an evaporator	• Potential reaction between TBP and UNH (red oil explosion) in a product concentrator • Overconcentration of U/Pu product (potential criticality) • Boil-over (release of radioactive materials from primary containment)
Waste Evaporation	• Overconcentration of waste	• Ruthenium escapes to stack • Leaks (to sumps) • Transfer error in the evaporator system • Coil or tube-bundle (in reboilers and condensers) failure	• Boil-over (release of radioactive materials from primary containment) • Hydrogen explosion in high-activity waste evaporator

Table 4-1 Potential incidents at a spent fuel reprocessing facility [DP-1558]

Process	Low Consequence	Intermediate Consequence	High Consequence
		• Overflow of a feed or bottom tank • High radioactivity in high-activity waste evaporator condenser • Inadequate vessel cooling capacity that leads to release of aerosols to room atmosphere • Leak of high-level waste concentrate	
Acid Recovery Operation	• Pressurization of acid absorber/fractionator	• Leaks due to corrosion • Eructation due to foreign materials in feed • Solvent addition to feed tank	• Red oil explosion due to solvent in feed or temperature > 140 ˚C • High radioactivity in recycled acid and/or water
Recovery Ion Exchanger Operation		• Overflow • Leakage due to corrosion	• Uncontrolled reaction between nitric acid and anion exchange resin in the primary recovery column • Ion exchange resin fire (leads to radioactivity release to environment)
Off-Gas Treatment Operation	• Loss of off-gas header volume • Iodine removal inadequate (increased iodine accumulation in krypton solvent recovery) • Excess nitrogen oxides in VOG iodine adsorber stream	• High radioactive particulate releases to building ventilation filters (increase release to sand filter) • Off-gas heater inadequately heats streams to iodine adsorbers • High ruthenium adsorber bed temperature • High krypton-85 releases	• Filter failure • Process vent system pressurized (leads to release to sand filter and smaller amount to atmosphere
Uranyl Nitrate Operation	• Overflow from storage tank • High uranium concentration in recovered acid	• Plugging of instrument lines and sensors during evaporation • Steam coil leak in evaporator reboiler • Cooling coil leak due to corrosion • Overflow in concentration system	
Uranium Denitration and Reduction Operation	• Pressurization of denitrator • High uranium concentration in recovered acid	• Over-pressurization of reductor • Fire in ammonia dissociator cubicle • Reductor malfunction which leads to release of UO_2 and hydrogen	• Denitrator eructation (leads to fire and explosions). Red oil explosion can occur in a denitrator if TBP is present. • Hydrogen explosion (hydrogen leaked from reductor)
Uranium Hexafluoride Operation	• Uranyl nitrate solution leaks • Uranium hexafluoride release • Airborne uranium oxide		• Criticality due to excessive uranium accumulation (high enrichment fuel) • Fire (due to solvent in feed, hydrogen leak and

Table 4-1 Potential incidents at a spent fuel reprocessing facility [DP-1558]

Process	Low Consequence	Intermediate Consequence	High Consequence
			presence of pyrophoric oxide powders)
Co-conversion Process Operation (UO$_2$-PuO$_2$ production)	• Transfer errors • Process solution leak • Overflows • Chemical addition error	• Pressurization of calciner • Calciner breached from internal corrosion • Calciner breached from impact (crane drop) • Excessive penetration of calcine through the calciner primary filter • Filter system breached • Fire suppression system failure • Uranium in steam condensate or cooling water returns the uranium oxide dissolver • Loss of power	• Ammonium nitrate explosion in calciner • Hydrogen explosion in product storage due to leak in hydrogen delivery system • Hydrogen explosion in Pu nitrate storage tank due to radiolysis • Uncontrolled reactions • Fire in process room • Hydrogen explosion in cold chemical area
Waste Calcination Operation		• Excessive penetration of calcine through sintered metal filters	• High temperature breach of the calciner • Calciner breached from internal corrosion, thermal shock, pressurization and impact • Filter systems breached
Solid Waste Processing Operation	• Airborne cement dust in grout mixer area • Radioactive contamination in cement preparation area • Failure of contaminated process components • Drums improperly filled (no cement added)	• Violation of stack release guide • Container failure • Power failure	• Fire in fuel hardware fixation area • Drum overflow in fuel hardware fixation area • Excessive fissile material in hulls • Waste container failure after filling • Fire in β-γ waste facility • Criticality potential in α waste
Solidification of Intermediate-Level Liquid Waste	• Transfer error that leads to high activity waste • Overexposure of personnel to radiation • Mixer plugging, which leads to worker exposure	• Airborne activity due to leaks, overflows, foam-out from mixer, suck back, and loss of vessel ventilation (power failure) • Waste container failure due to weld failure, fault in drum, corrosion, or impact	• Fissile material in feed • Uncontrolled reaction in mixer or product container
Vitrified High-Level Waste Storage Operation	• Contaminated canisters • Water loss from storage pool	• Loss of cooling water and shielding • Canister stress corrosion • Canisters raised above adequate shield level • Canisters dropped during handling • Waste transfer line rupture due to the high viscosity sludge causing plugging,	• Fire

Table 4-1 Potential incidents at a spent fuel reprocessing facility [DP-1558]

Process	Low Consequence	Intermediate Consequence	High Consequence
		leading to potential accumulation of flammable gases and heating of the decay heat in high-level waste [Jain 1998] • Electrically heated melter short due to accumulation of noble metal on melter floor [Jain 1998] • Waste pretreatment issues that lead to release of Pu with the effluent in the evaporator [Jain 1998] • Improper use of chemicals in feed preparation that leads to erosion and corrosion of on-line instruments, causing malfunctions of on-line pressure, temperature and level instruments [Jain 1998]	

[1]During the head-end process when the spent fuel rod is being chopper or crushed, magnesium-clad fuel and uranium/plutonium metal fuel are exposed to air, meaning that combustion may occur at normal temperatures [OECD 2005]. Zircaloy debris or dust could pose a potential fire or explosion if this material is used for the cladding [IAEA DS360].
[2]N-dodecane in TBP in the PUREX process poses potential combustion conditions [Ikeda 2003].

Table 4-2   Summary of possible major accidents at a spent nuclear fuel reprocessing facility

Accident	Cause	Consequence
Spill		
Rupture of a high-level liquid waste storage tank	• Corrosion and erosion • Mechanical stress from overpressure and loss of cooling • Simultaneous failure of the vessel off gas (VOG) system (e.g., by a H_2 explosion from loss of air sparge)	• Release of tank contents to room floor • Airborne release of tank contents in cell • Potential for large release to process stack if safety feature fails • Decay heat in the released liquid may cause any liquid to boil away and melts the remaining solids that can cause additional gases and airborne aerosols
Fire		
Solvent fires	• High temperature in organic streams • Solvent leak and vaporization • Solvent sprays on hot equipment (e.g., steam line, evaporator reboiler) • Solvent spill from skimmer overflow, flooded decanter, and makeup tank overflow • Ignition source present • Room atmosphere oxygen concentration above limit	• Airborne activity in process room • Possible overheating and plugging room ventilation filters • Air reversal • Loss of process control • Equipment damage • Energy release by solvent burning somewhat limited by amount in the process room

Table 4-2   Summary of possible major accidents at a spent nuclear fuel reprocessing facility

Accident	Cause	Consequence
Ion exchange resin fire or explosion	• Self-heating of resin due to abnormal conditions in column: high nitric acid concentration (9 molar [NUREG-1909]), column overloading, dry resin in column, and high column temperature • Self-heating of spent resin in waste • Spontaneous combustion of spilled resins • Exposure of resin to strong oxidants other than nitric acid, such as permanganate or chromate ions, to high radiation doses, to strong reducing agents such as hydrazine and to catalytic metals such as iron, copper or chromium [NUREG-1909] • Allowing resin to remain in a stagnant, non-flow condition while loaded with exchanged metal and/or in contact with process concentrations of nitric acid [NUREG-1909]	• Column pressurization • Column rupture • Eruption • Airborne activity in room, release of fission products to ventilation system • Possible plugging or overheating of VOG filters • Waste fire • Room pressurization
Hydrogen Explosion		
Feed tank or vessel vent system	• Concentration of feed solution • Hydrogen produced by radiolysis of feed solution • Loss of purge to feed tank • Oxygen source • Ignition source	• Severe damage to equipment • Potential damage to off-gas system • Release of radioactive material to process room • Potential initiation of fire
High activity feed, plutonium evaporator bottom tank, product storage tank, mixed-oxide calciner	• Hydrogen produced by radiolysis of product solutions • Ignition source • Loss of vessel purge	• Damage to off-gas system and release of off gas • Equipment damage • Release of fissile to room floor • Room pressurization
VOG header	• High H_2 or HT content in VOG stream, air in stream, and ignition source • Propagation from explosions in other equipment in room	• Aerosol release by filters • Damage to VOG system • Pressurization of process room via room exhaust header
Process room	• Ignition presence • Excessive oxygen presence in the room atmosphere	• Damage to shielding room, ventilation and VOG systems
Other Explosion[1]		
Red oil explosion in concentrator, uranium evaporator (denitrator)	• Solvent in feed • Temperature > 130 ˚C • Heavy metal (U, Pu) present	• Rupture of concentrator may cause violent ejection of liquid with formation of aerosol • Damage to VOG treatment system, leading to the release through stack. • Induced fire • Projectile generation in room • Room pressurization

Table 4-2   **Summary of possible major accidents at a spent nuclear fuel reprocessing facility**

Accident	Cause	Consequence
Hydrazoic acid explosion	• Uncontrolled reaction of hydrazine products used in solvent recovery. Hydrazine is used in the extraction step during the Pu stripping operations	• Damage to VOG system • Fire in solvent recovery • Room pressurization • Airborne activity
Criticality		
Fuel storage	• Distortion of fuel storage array • Fuel improperly stored • Fuel assembly dropped into fuel storage array • Fissile material on pool water filter	• Damaged elements could release short-lived noble gases and iodine • Radiation and neutron locally high but largely shielded by pool water
Inadequate poison in solvent	• Poison concentration too weak • Wrong chemical added • Failure to add poison at correct volume	• Dissolver (in conjunction with undetected plugging of liquid or solids) could release volatile fission products Digester (in conjunction with undetected accumulations of solids or with over-concentrated solution) could release volatile fission products
In mechanical processing and feed preparation operations	• Inadequate poison in solvent • Overconcentration of solution in digester or feed adjustment, followed by precipitation • Accumulation of fissile residue in digester or in solids recycle tank • Voloxidizer flooded with water • Caustic added to feed • Dissolver blockage in addition to loss of poison • Accumulation of chopped fuel on undetected stuck-shut voloxidizer and dissolver	• Criticality potential in feed tanks • Probable release of airborne activity, including noble gases and iodine to room atmosphere and off-site system • Possible severe equipment damage • High radiation levels in rooms. • Mass and energy probably contained in room • Energy release on the order of 10^{18} fissions
Solvent extraction operations	• Excessive fissile material in high activity centrifuge bowl • Fissile material in unintended location • Damage to equipment • Pu reflux in first, second, or plutonium purification cycle • Pu precipitation • Pu over-evaporation • Fissile uranium reflux • Fissile material in solvent tank	• High local radiation • Release of gaseous fission products to room or to VOG • Damage to equipment
In miscellaneous systems	• General purpose concentrator (GPC) bottoms-caustic routed from rework decanter to digester in mechanical process; plutonium precipitation • GPC bottoms routed from rework decanter to high-level aqueous waste concentrator • Fissile material in high activity waste; plutonium precipitation • Excessive fissile material accumulation in organic phase in rework decanter	• High local radiation levels • Release of gaseous fission products to room and VOG systems • Damage to process equipment; possible loss of process control
Loss of Coolant		

Table 4-2   Summary of possible major accidents at a spent nuclear fuel reprocessing facility

Accident	Cause	Consequence
	• Loss of power or other equipment failure can cause a loss of coolant to maintain temperature in a high-level waste (HLW) tank. • Heating of the HLW tank leads to evaporation of solvents and radioactive material. It can also lead to chemical instabilities or fires.	• Release of fission products and other isotopes from the HLW tank in the form of vapors and aerosols • High local radiation levels • Potential release through off-gas systems into the atmosphere • Possible damage to process equipment from fire

*This summary is obtained from [NUREG-1320] but was further condensed for this report. The accident and consequence information originated from [DP-1558]. Additional information is also provided in the indicated references.

[1]An explosion due to the decomposition products of hydrazine, hydrazoic acid (HN_3) and nitrates is also possible. The formation of these chemicals depends on the concentration of technetium and the flow of plutonium in the PUREX process [OECD 2005]. Equipment that can produce red oil include evaporators (these evaporate the diluted aqueous process streams, mainly metal nitrates, and are used to boiling away volatile water and nitric acid components), acid concentrators (these distill the acid solutions), and denitrators (these are known as calciners, which are heating devices, that heat concentrated solutions of metal nitrates to the point of decomposition) [DNFSB_T33].

According to a recent study [BNL 2011], analytical approaches for hazard analysis have been developed for reprocessing facilities that include methods of hazard analysis for chemical plants. These methods are employed to identify, model, and quantify failure frequencies of individual processes or the entire facility. Information about these quantifications and models is described in [BNL 2011]. Table 4-3 lists potential accident initiators.

Additional sections describe each type of accident in more detail. Since chemical explosions are highly energetic, these are discussed first, followed by inadvertent criticality, then fire, and finally spill accidents. In each of these sections, brief descriptions are provided for previous accidents and incidents. Finally, a discussion is provided regarding the phenomena related to the generation of airborne material.

Table 4-3 Potential accident initiators from external events and natural phenomena [IAEA DS360]

Accident Initiator	Description
External Event	
Plane crash	If the facility is located near an airport, there is a potential for an airplane to crash into the facility. The probability of a plane crash is greatly dependent on the footprint of the facility and the distance from the airport. The over-flight impact probability is much smaller than impacts when the plane is landing or taking off.
Vehicle accidents	If there is inadequate distance from the facility to roads or no means of protecting the facility from vehicle accidents, there would be a potential of vehicle accidents that may impact the facility.
Explosion at nearby facilities	If a nearby facility can produce an explosion that can impact the facility of interest, then there is an explosion concern for the facility of interest. These concerns include explosion fragments flying into the facility and/or significant overpressure that could impact the operations of the facility. For example, a blast may rupture a solution tank which leads to a spill of radioactive materials, fire or explosion.
Natural Phenomena	
Earthquakes	Without seismic qualified equipment and containers used in the facility, seismic-induced fire and explosions are possible.
Severe weather conditions	Wind, rain, lightning, snow, and extreme temperatures are identified as severe weather conditions. If the roof was not designed to withstand the design loads of winds, rain, and snow, disruption of the operations within the facility may be possible. Lightning protection design and adequate ventilation can deal with lightning strikes and extreme temperature changes, respectively. Temperature is very important because many of the chemical processes used in the fuel reprocessing facility require a certain range of operating temperature. Without this control, fire and/or explosions due to elevated temperature may be possible.

4.1   Chemical Explosions

This section presents an overview of the historical explosion accidents at reprocessing facilities and discusses the pertinent phenomena associated with chemical explosions in Section 4.1.1 and Section 4.1.2, respectively.

4.1.1   History

As described in the introduction, both fires and explosions are potential accidents because of the corrosive and reactive chemicals used in the extraction and purification phases in fuel reprocessing. Both the temperature and molarity of the chemicals in the process phases must be controlled to prevent conditions conducive for fires and/or explosions. Table 4-4 lists explosion accidents that have occurred throughout the history of reprocessing facility operations along with a description of the events that led to the explosions. As shown in Table 4-4, the dominant historical explosive event has been attributed to red oil. Chemical explosions can also result from hydroxylamine nitrate (HAN) mixtures with nitric acid. Hydroxylamine sulfate (HAS) has similar properties similar to HAN. Both have been used in the PUREX process in extraction and purification operations. An explosion of approximately 75 tons of TNT equivalent is the largest explosion in a reprocessing facility that has been reported (see Table 4-4).

Table 4-4 Explosion accidents in fuel reprocessing plants

Facility	Year	Accident	
		Type	Description
Hanford, WA, U.S. [DNFSB_T33]	1953	Nitration oxidation reaction – equipment failure	The accident occurred in Building 321 where a feed pump failed during the initial operation of a new evaporator using fresh uranium. TBP was inadvertently present in the feed. Despite the pump failure, the evaporation process continued causing normal concentrations and temperatures to be exceeded. A great deal of red fumes escaped leading to an explosion.
Savannah River Site, SC, U.S. [DNFSB_T33]	1953	Nitration oxidation reaction – equipment failure/ operator error	A UN* solution was being batch-concentrated in the TNX Facility to remove excess nitric acid from solution. The UN was in contact with 30% TBP solvent in kerosene. Approximately 80 lbs. of TBP was inadvertently present in the feed, causing the temperature to rise, which ultimately led to an explosion.
Mayak Production Association, Russia [Todd 2008]	1957	Nitration oxidation reaction	Liquid high-level waste containing sodium nitrate and acetate salts from the acetate precipitation process was stored in underground tanks. The cooling system in one of the tanks failed, and the temperature in the tank rose to 350 °C. The tank contents evaporated to dryness, causing a massive explosion equivalent to 75 tons of TNT. Over 20 million Ci of radioactivity was released to the environment.
La Hague, France [NUREG-1909], [Scheider 2001]	1970	Explosion	The temperature of the dissolution reaction of the graphite fuel increased sharply, resulting in hydrogen generation, which led to an explosion and release of fission products. 5900 GBq was recorded at the filter of the stack, mainly due to Sb-125 (95%) and I-131 (5%) [Scheider 2001]
Windscale, U.K. [NUREG-1909]	1973	Fire/explosion	An exothermic chemical reaction in a reprocessing tank.
Savannah River Site, SC, U.S. [DNFSB_T33]	1975	Nitration oxidation reaction – equipment failure/ human error	At the H-Canyon Outside Facilities, UN was being calcined to UO_3 along with an unknown quantity of organic material. Prior to the explosion, dense red fumes were emitted into the denitrator room.
Hanford [Cadwallader 2005]	1976	Explosion	An ion exchange column suffered a chemical reaction and explosion. A worker was in front of the glovebox that housed the column at the time of the explosion. The worker was injured by glass from the glovebox's shattered window, and the worker received Americium-241 contamination as well. The americium had been collected on the ion exchange resin before the explosion and was dispersed throughout the room in the course of the explosion.
Idaho [Cadwallader 2005]	1991	Explosion	On February 9, 1991, a small explosion occurred at the Idaho Chemical Processing Plant that ruptured a 6-inch diameter borosilicate glass vessel during dissolving operations to retrieve 1.9-kg of highly enriched, unirradiated uranium fuel from a scrap fuel can of predominantly uranium-zirconium. The uranium was contaminated with thorium, plutonium, and other nuclides. The dissolver vessel was set up within a walk-in confinement hood but in the course of the explosion, some of the hot nitric acid and uranium was splattered onto three operations personnel in the area. They were not injured by the acid or by glass shards but the three operators and one emergency response worker were alpha contaminated; the contamination was not significant. The second dissolver, next to the failed unit, was intact and there was no damage to other equipment. Evidence suggested that a reaction of epsilon phase $U\text{-}Zr_3$ alloy and nitric acid occurred.

Table 4-4 Explosion accidents in fuel reprocessing plants

Facility	Year	Accident	
		Type	Description
Mayak Production Association, Russia [NUREG-1909]	1993	Fire/explosion	A tank containing a solution of paraffin hydrocarbon and TBP exploded. The explosion was strong enough to knock down walls on two floors of the facility and caused a fire. According to [IAEA 1998], the accident caused damage to both the reprocessing line and the building. This resulted in the release of about 30 TBq (810 Ci) of beta and gamma-emitting radionuclides and about 6 GBq (0.162 Ci) of ^{239}Pu.
Tokai, Japan [Todd 2008]	1997	Fire /explosion – operator error	A fire occurred in the bitumen waste facility of the reprocessing plant. Bitumen is used to solidify intermediate-level liquid radioactive waste. The fire apparently occurred after errors were made in the monitoring of a chemical reaction. The fire was not completely extinguished and about ten hours after the chemicals had accumulated, an explosion occurred which failed the confinement of the facility.
Hanford, WA, U.S. [Todd 2008] [DOE/EH-0555]	1997	Explosion – operator error	A storage tank containing HAN and nitric acid was allowed to evaporate to dryness. The resulting explosion destroyed the tank and blew a hole in the roof of the building.

*UN is uranyl nitrate

The formation of a nitration oxidation reaction can be explained as follows [IRSN 2008]. The organic solution of tributyl phosphate (TBP), diluents (such as kerosene, n-dodecane, hydrogenated tetra-propylene), and the products of their degradation by hydrolysis and radiolysis can react violently with oxidizing products. At high temperatures, these solvents in the presence of nitric acid or heavy metal nitrates (such as plutonium nitrate and uranyl nitrate) can lead to the formation of what has been traditionally called red oil. Red oil is a substance of non-specific composition formed when an organic such as TBP and kerosene comes in contact with concentrated nitric acid and is heated above 120 °C under reflux. The red oil is the result of formation of nitrated organics. In the PUREX process, this red oil can be formed in evaporators that concentrate aqueous flows containing nitrates and traces of organic solvents. These flow streams include (a) "inter-cycle" concentrating of uranyl nitrate solutions; (b) concentrating fission products; (c) acid recuperation, where the raffinates from various extraction cycles are concentrated; and (d) effluent stream flow after the oxalate precipitation of plutonium. Red oil formation requires prolonged heating and involves complex processes such as radiolysis, hydrolysis, oxidation, and nitration reactions. In addition, red oil formation depends not only on the level of nitrates and temperature of the medium, but also on the level of radiolytic degradation of the solvent.

In French reprocessing facilities, the risk of a nitration oxidation reaction is mainly associated with the following evaporator operations [IRSN 2008]:

- Inter-cycle concentration of re-extraction solutions of uranium from the first extraction cycle. The first extraction cycle is composed of three stages: co-extraction of uranium and plutonium, followed by selective partitioning of plutonium, and then uranium partitioning before spent solvent treatment.

- Concentration of fission product solutions from the first extraction cycle.

- Recovery of tritiated and non-tritiated acids where aqueous acid solutions of low and medium activity are concentrated.

- Treatment of the OML before recycling the upstream phase of oxalic plutonium precipitation.

- Treatment of organic effluents by distillation.

Based on the lessons-learned from nitration oxidation reactions at DOE facilities, the following controls have been identified to mitigate or prevent a nitration oxidation reaction [DNFSB_T33]:

1. Maintaining a temperature of less than 130°C to prevent explosion conditions. This means adequate cooling is required.

2. Process equipment/vessel design that includes sufficient venting to prevent an overpressure event from destroying the equipment/vessel.

3. Decanters or other liquid-liquid separation equipment that may be used to remove excess TBP from feed streams entering the heated process equipment (such as evaporators and denitrators [NUREG-1909]).

4. Limiting the process equipment/vessel size and organic concentration to to restrict explosive energy to be low enough to mitigate this type of accident.

5. Limiting the nitric acid concentration to less than 10 molar.

4.1.2 General Discussion

This section provides a general discussion of the phenomenological effects associated with an explosion. An explosion event in a fuel reprocessing facility begins with an increase in mass or energy that produces a stress load upon the equipment barrier. If the load is sufficient, a failure of the process equipment barrier can occur. The pressurization event could result from chemical reactions due to equipment or control system failures, temperature regulation failure, relief valve failures, etc.
Either as a consequence of the vessel depressurizing or the kinetics of the energetic event, a pressure pulse is generated and imparts a force on the constituents of the reprocessing component such as the liquid or solid contents. The force imparted by the pressure wave can overcome liquid adhesion and gravitational forces, effectively separating the fluid body into suspended droplets. Coupled with a failure of a vessel, a portion of the newly suspended material may partition to the vapor space of the surrounding atmosphere.

The initially suspended mass is a distribution of various droplet sizes. The size distribution is dependent on the energy of the event, the density of the susceptible material, evaporation from the material, etc. Proper characterization of the droplet size distribution permits meaningful determination of effective removal processes that would reduce the ultimate airborne material released to the environment.

A threshold aerosol diameter may be applied to simplify the calculation space since gravitational settling is the overwhelming deposition mechanism for larger particulates. Particles with an aerodynamic diameter of 50 micrometers or less can be categorized as an aerosol. Aerosol deposition mechanisms such as diffusiophoresis, Brownian motion, and thermophoresis become relevant deposition processes much below this 50 micrometer threshold. Effects on larger droplets such as evaporation, impacting and splattering are phenomena that may further increase the fraction of airborne material considered to be aerosolized.

In addition to the deposition mechanics, aerosols interact directly with one another. Again, based on the size of the aerosol, various agglomeration mechanisms occur at different rates. Agglomeration due to the following mechanisms must be considered: Brownian motion, gravitational settling, and shear and inertial turbulence.

Similar to the release of liquids, vapors within the process equipment may be available for transport as a consequence of the equipment failure. In addition to any direct release of vapor, evaporation from the surface of the remaining fluid within the vessel, content spilled from the vessel, and airborne products will each contribute to the generation of vapor species resulting from an explosion. Condensation of vapor species can reduce overall availability to contribute to the environmental source term. Vapor condensation onto structures, pool surfaces, and aerosols present may occur, each of which affects the total airborne material.

An explosion may damage more than the equipment or space where the event originated.   Any remaining energy from the explosion (in the form of a pressure pulse) is imparted to the surrounding walls and ceiling. Once the explosion occurs in a vessel or container, the overpressure may further damage the surrounding walls and ceiling in a room. The propagation of the explosion greatly depends on the geometry of the room and the connecting rooms, and the initial explosion location. A determination of damage to the subsequent connecting rooms and near field equipment could indicate possible cascading failures resulting from the initiating explosion.

Typical blast overpressures for a given explosion are given in Table 4-5. This table provides the differential pressure (or overpressure) that may be required to cause damage to common structures. Typically, the retaining walls used in the fuel reprocessing facilities are thick to shield intensive radiation emanating from the radioactive spent fuel materials. The shielding thickness can be as high as several feet of concrete with rebar reinforcement and may include a steel liner. The ceiling of the fuel reprocessing facilities, particularly in the area of the chemical processing areas, may be constructed to accommodate the ventilation and other piping systems above it. Thus, the ceiling could possibly withstand overpressure but may not be as strong as surrounding walls made from reinforced concrete.

A more detailed discussion on the blast overpressure and the failure models is presented in Section 5.2.1.4.

Table 4-5 Blast overpressure on structural damage [Merrifield 1990]

Pressure Differential		Damage
(psig)	(kPa)	
0.15	102.359	Typical pressure for glass failure
0.4	104.083	Limited minor structural damage
0.5-1	104.772-108.220	Large and small windows usually shattered, occasional damage to window frames
0.9	107.530	Roof damage to oil storage tanks
1.3	110.288	Steel frame of clad buildings slightly distorted
1.5	111.667	Slight damage to window frames and doors
2-2.5	115.114-118.562	Some frame distortion of steel framed buildings
2-3	115.114-122.009	Concrete or cinder brick walls 8" (20.32 cm) to 12" (30.48 cm) thick, not reinforced shattered
3-4	122.009-128.904	Rupture of oil storage tanks
4	128.904	Cladding of light industrial buildings ruptured
4.5	132.351	Severe distortion to frames of steel girder framed buildings
7-9	149.588-163.378	Collapse of steel girder famed buildings
8-10	156.483-170.272	Brick walls completely demolished
>10	>170.272	Complete destruction of all unreinforced buildings
13	190.957	18" (45.72 cm) brick walls completely destroyed
280	2031.857	Lip of crater

4.2 Inadvertent Criticalities

This section presents an overview of the historical criticality accidents at reprocessing facilities and discusses the generalities of criticality accidents in Section 4.2.1 and Section 4.2.2, respectively.

Much of the information presented in this section is adapted from LA-13638 [McLaughlin 2000]. Similar criticality accidents are discussed in [Todd 2008].

4.2.1 History

As of the year 2000, there have been a total of 22 major process-related criticality incidents in the world [McLaughlin 2000]. Figure 4-1 shows the chronology of these 22 accidents These accidents can be summarized as follows:

- 21 of 22 excursions occurred in solutions or slurries.

- 18 accidents occurred in manned and unshielded facilities.

- Nine fatalities resulted from these accidents.

- No equipment was damaged in these accidents.

- One accident resulted in measurable fission product contamination beyond the plant boundary and in measurable exposures to the public at Tokai Fuel Fabrication Plant.
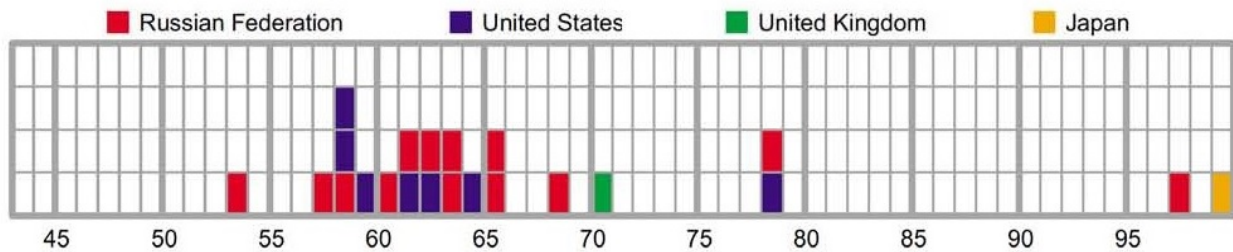
Figure 4-1   Chronology of process criticality accidents. Plot shows number and location of accidents by year from 1945-2000. [McLaughlin 2000]

Among these incidents, one is related to metal ingots, which is not included in further discussions. Additionally, not included in further discussions are process accidents related to enrichment and uranium hexafluoride such as:

1.   Siberian Chemical Combine, July 14, 1961 – Accident occurred in a gaseous diffusion uranium enrichment facility.

2.   Electrostal Machine Building Plant, November 3, 1965 – Accident related to conversion of the uranium hexafluoride to uranium oxide.

3.   Siberian Chemical Combine, December 13, 1978 – Accident related to plutonium metal ingots in a storage container.

4.   Novosibirsk Chemical Concentration Plant, May 15, 1997 – Accident related to chemical etching process in the fuel rod production.

Table 4-6 provides the remaining criticality accidents that can be related to fuel reprocessing facilities. As shown in this table, the location of the accident, cause, accident description, and total fissions of the accidents are identified.

Table 4-6 A summary of solution criticality incidents [McLaughlin 2000]

(#)Accident	Operation [Cause]	Description	Total Fissions
(1) 3/15/1953, Mayak Production Association, Russia	Plutonium purification [operator error/procedural error]	There were 18 vessels containing plutonium nitrate solution in a staging area where operations of mixing, dilution, volume measurement, sampling, and interim storage were done. Each vessel had a 500 g plutonium mass limit. An excursion occurred in Vessel 18 (originally contained 26 liters), which contained 848±45 g of plutonium in 31 liters at the time of the accident. The cause of this excursion was due to unrecorded transfer of 5 liters of solution from Vessel 1 to Vessel 18.	2×10^{17} fissions. This was based on a temperature rise of 60 ˚C in 31 liters of the solution.
(2) 4/21/1957, Mayak Production Association, Russia	Uranium purification [equipment failure – defect in the filter fabric]	Impure uranyl nitrate (90% enriched uranium) and oxalic acid solution were mixed in a vessel to precipitate the uranyl oxalate trihydrate, which was done within a glovebox. This precipitate slurry was then vacuumed to a holding tank from which it was drained into a filter vessel. The precipitate containing uranium was collected on the filter fabric, and the filtrate was pulled through by vacuum and collected in a filtrate receiving vessel. An excursion occurred in this vessel that caused the filter vessel to bulge upward, followed by a violent release of gas and ejection of some of the precipitate out of the filter vessel and onto the glovebox floor.	A fission yield of 1×10^{17} fissions. This yield was based on the measurement in the room and the concentration of ^{24}Na in the operator's blood (17 hours after the accident, the dose was 245 Bq/cc).
(3) 1/2/1958, Mayak, Production Association, Russia	Solution critical experiment [operator error]	Based on the 4/21/1957 accident, a solution criticality experiment was conducted using highly enriched uranyl nitrate solution (90% enriched). The experiment was conducted in a cylindrical vessel that could hold an excess of 400 liters of solution to measure a wide range of critical states. Typically after each experiment, the solution must be drained to a smaller vessel which was criticality-safe. Experimenters decided one day to circumvent the routine by manually pouring the solution out of the experiment vessel. This routine created an excursion because human bodies provided additional reflection of the neutrons, which created a criticality-unsafe configuration.	Based on the fission product activity in the solution, a single pulse event was about 2×10^{17} fissions.
(4) 6/16/1958, Y-12 Plant, Oak Ridge, TN, U.S.	Uranium recovery [equipment failure]	Solid wastes with highly enriched uranium (93% enriched) were dissolved in nitric acid, purified, concentrated, and then converted to uranium tetrafluoride. A 55-gallon drum served as a residue collection tank. Because there were apparent leaks in the system, liquid accumulated in the drum to about ~56 liters with 2.1 kg of^{235}U mass. This condition yielded excursions that lasted about 20 minutes.	Based on the instrument at the site and detectors nearby, the first spike was about 6×10^{16} fissions, with a total of 1.3×10^{18} fissions.
(5) 12/30/1958, Los Alamos, NM, U.S.	Plutonium recovery [equipment design]	An organic treatment tank, which typically contained solutions < 0.1 g Pu/liter and traces of americium. This tank was made of stainless steel and had a volume of 1000 liters. Before the accident, the tank contained an aqueous layer of 330 liters with 60 g of Pu, and an organic layer of 160 liters with 3.1 kg of Pu. The	Based on the reactivity of the system, the excursion yielded 1.5×10^{17} fissions.

Table 4-6 A summary of solution criticality incidents [McLaughlin 2000]

(#)Accident	Operation [Cause]	Description	Total Fissions
		aqueous layer was located at the bottom of the tank. This configuration was subcritical. When the stirrer was started, the motion altered the reactivity of the tank, and an excursion occurred in about 3 seconds. The excursion was terminated by the stirring action. After this accident, a criticality-safe tank was used.	
(6) 10/16/1959, Idaho Chemical Processing Plant, ID, U.S.	Uranium extraction [operator error, equipment failure]	The impurity extraction process occurs in three pulse columns. Intermediate between the first and second cycle extraction, the uranyl nitrate solution (91% enriched uranium) was stored in two banks of long pipes (referred to as pencil tanks). There was a line leading from the interconnected pipes to the 18,900 liter waste receiving tank. As required, operators needed to conduct analysis by collecting samples using a sparging operation. Because the air sparging was so forceful, and the pressure gauge of the other pipe was functioning improperly, the liquid in the pipes was forced to the top of the loop that drained into the receiving tank. Excursions boiled away half of the 800 liter solution volume that terminated the criticality.	Based on the strip chart recordings from the continuous air monitors at various locations, the initial spike of at least 10^{17} fissions, followed by boiling of 15 to 20 minutes to yield a total of 4×10^{19} fissions.
(7) 12/5/1960, Mayak Production Association, Russia	Plutonium recovery [operator error]	The accident occurred in the waste solution process to recover plutonium. A total of 4 purification vessels were used in the process. All vessels were contained in glove boxes. A holding tank was used to keep solutions for various stages of the purification process. Because of overloading, the mass limit in the initial purification process was passed, and subsequent purification process resulted in a criticality accident. Excursions occurred in the holding tank, which contained 894 g of plutonium. 180 g of Pu was flushed out of the holding tank during the accident.	The excursions yielded a total of 2.5×10^{17} fissions.
(8) 1/25/1961, Idaho Chemical Processing Plant, ID, U.S.	Uranium purification [operator error, equipment design]	The accident occurred in the upper disengagement head of the H-110 product evaporator. Because the disengagement head was not a favorable geometry, any solution that reached this section may yield criticality. Despite an overflow line to prevent a significant amount of solution reaching this section, 200 g of uranyl nitrate solution with 90% enrichment per liter was apparently rapidly ejected up into this section causing excursions. This action was due to operators attempting to clear the plugged line with air. The cause of the ejection of the solution upward was due to the high air pressure which forced the solutions in the favorable geometry section to move upward into the disengagement head.	Because of no instrument readout of the event, it was calculated to be 6×10^{17} fissions.
(9) 7/14/1962, Siberian Chemical Combine, Russia	Uranium enrichment plant [equipment	The accident was caused by the accumulation of high concentration of UF_6 in the vacuum pump oil reservoir. Four days before the accident, there was a sudden flow rate increase of UF_6	The total number of fissions for both excursions was estimated by fission

Table 4-6 A summary of solution criticality incidents [McLaughlin 2000]

(#)Accident	Operation [Cause]	Description	Total Fissions
	failure and operator error]	into the reservoir The operator turned on the vacuum pump and approached the equipment (~0.5 m) to open a valve between the pump and holding vessels. This resulted in a criticality event.	product analysis to be about 1.2×10^{15}.
(9) 4/7/1962, Hanford Works, U.S.	Plutonium recovery [operator error]	The transfer tank with a capacity of 69 liters received solution containing 45 g Pu per liter and later additions of other diluted solutions which caused an excursion that lasted for 37.5 hours. The shutdown mechanism for the accident was due to a central pipe that entered the bottom of the vessel. Fission reactions may have driven off CCl_4 and converted most of the remaining organic to dibutyl phosphate. This phosphate, with extracted plutonium, may have gone to the bottom of the vessel and into the pipe to stop the excursion.	An initial spike estimated to be no more than 10^{16} fissions, and lasted for 37.5 hours to total fissions of 8×10^{17}.
(10) 9/7/1962, Mayak Production Association, Russia	Plutonium recovery [operator error and equipment design]	Dissolution vessels were used to dissolve solid Pu residue to further purify plutonium. Due to the unfavorable geometry of the vessels and undetermined plutonium concentration, excursions occurred in the vessels.	A total of three excursions were detected and estimated to have total of 2×10^{17} fissions.
(11) 1/30/1963, Siberian chemical Combine (Tomsk), Russia	Uranium recovery [operator error]	The recovery process required that dry precipitate of uranium (90% enriched) be placed into concentrated nitric acid for the dissolution process. The criticality control was based on the total fissile mass and fissile concentration. Because the operator recorded the concentration of the uranium precipitate incorrectly, this action led to the first excursion in a large diameter collection vessel. Then for the next 10 hours, eight additional excursions occurred.	The total fissions was estimated to be 7.9×10^{17}.
(12) 12/2/1963, Siberian Chemical Combine (Tomsk), Russia	Uranium purification [equipment design]	A number of process vessels containing both aqueous and organic solutions of uranium (90% enriched) were used for the purification process. In each vessel, a level indicator showed the level of the solution. However, this indicator worked well with aqueous solutions, but not with organic solutions. Since the organic solution is less dense than an aqueous solution, the organic solution was transferred to other vessel via the vacuum system. Over time, a final vessel accumulated excessive organic solution which led to excursions, which lasted for hours.	Based on the ^{140}La fission product analysis, the total fissions was about 6×10^{16}.
(13) 7/24/1964, United Nuclear Fuel Recovery Plant (Wood River), U.S.	Uranium recovery [operator error]	An operator mistook a highly concentrated uranyl (93% enriched uranium) nitrate solution as trichloroethane as the washing fluid for the solvent extraction columns. The operator pulled this solution into a vessel containing a large quantity (41 liters) of sodium carbonate solution. A series of excursions occurred after the combined solution (about 51 liters) was stirred automatically.	The first excursion was estimated as 1.0×10^{17} to 1.1×10^{17}, with 20% of the combined solution ejected. The total fissions for the entire event were about 1.30×10^{17} to 1.55×10^{17}.

Table 4-6 A summary of solution criticality incidents [McLaughlin 2000]

(#)Accident	Operation [Cause]	Description	Total Fissions
(14) 12/16/1965, Mayak Production Association, Russia	Uranium recovery [equipment design]	The residues (90% enriched uranium) being recovered were produced from dissolution, precipitation, and reduction processes. The accident occurred in a dissolution vessel or holding vessel. A total of 11 excursions occurred. The cause of the accident was probably due to unfavorable critical configurations of the vessels.	The total fissions for the accident were about 5.5×10^{17}.
(15) 12/10/1968, Mayak Production Association, Russia	Plutonium processing [equipment design and operator error]	An unfavorable vessel geometry was used to store a plutonium organic solution. The operator was instructed to pour 35 liters of plutonium organic and aqueous solutions into a 60-liter vessel. A first excursion occurred with 16 liters containing 880 grams of plutonium ejected. The remaining plutonium in the vessel was about 709 grams.	The total fissions was about 1.3×10^{17} with the first excursion was about 3×10^{16}.
(16) 8/24/1970, Windscale Works, United Kingdom	Plutonium recovery [equipment design]	The plutonium recovery was done using a dissolver, conditioner, and transfer vessel. The criticality accident occurred in the transfer vessel, where excess plutonium solution accumulated, and was well above the criticality limit. A large mass of plutonium accumulated in the vessel over a period of 2 years.	The total fissions were about 10^{15}. It lasted to less than 10 seconds.
(17) 10/17/1978, Idaho Chemical Processing Plant, U.S.	Uranium extraction [equipment failure and operator error]	A pulsed column with larger diameter regions at the top and bottom of the column is used in the uranium solvent extraction phase. Two streams of organics (with a density lighter than aqueous solutions), and the aqueous solutions were flowing in the column. The organic solution containing uranium was extracted in the column. The accident was due to a number of causes. The first cause was the leaking water valve on the make-up tank. The chart recorder had run out of paper and procedures were not followed. The event was caused by the accumulation of the solution over time (~1 month).	Total fissions were about 2.7×10^{18}.
(18) 9/30/1999, JCO Fuel Fabrication Plant, Japan	Uranium processing [operator error]	A special request to process uranyl nitrate solution at 18.8 % enrichment of uranium was done for this operation. The operation had been done routinely for uranium enrichment of up to 6%. Manual pour of the solution from one vessel to another vessel was performed in the operation. To save time for dissolution, the operators did not follow the procedure, and poured the bucket of solutions to a precipitation vessel with an unfavorable geometry. Eventually excursions occurred.	Total fissions were about 2.5×10^{18}.

Based on these process accidents, the following lessons have been identified:

1.   Vessels with geometries that favor criticality should be avoided, particularly when high concentration solutions might be present.

2.   No verbal procedures should be used. Instead, documented procedures should be used.

3.   Operators should be aware of any abnormal conditions of the processes.

4.   Fissile material accountability must consider criticality control when tracking material accumulations in the equipment.

5.   Operators should be trained to respond to potential (based on past experience or PRA analysis) equipment failure or human errors.

6.   Operators should be trained to deal with possible off-normal conditions for the processes involving both organic and aqueous solutions.

7.   Operators should be trained to follow procedures.

8.   Operators should be trained to deal with failure of hardware that is important to criticality control.

Table 4-7 provides the single-parameter criticality safety limits that are often used to prevent inadvertent criticality events from happening. An inadvertent criticality event is unlikely to occur when one of these limits is used to control the operation. However, this is not always practical. For example, a spent fuel reprocessing facility that processes a large amount of uranium cannot be limited to the amount of fissile and fissionable materials given in Table 4-7. Therefore, criticality safety evaluations are often performed to determine the limits to prevent inadvertent criticality.

Table 4-7   Single-parameter limits for uniform aqueous solutions of fissile nuclides (U-235 and Pu-239) [ANS-8.1]

Parameter	Subcritical Upper Limit for Fissile Solute		
	$^{235}UO_2F_2$	$^{235}UO_2(NO_3)_2$	$^{239}Pu(NO_3)_4$
Mass of fissile nuclide, kg	0.76	0.78	0.48
Diameter of cylinder of solution, cm	13.7	14.4	15.4
Thickness of slab of solution, cm	4.4	4.9	5.5
Volume of solution, liter	5.5	6.2	7.3
Concentration of fissile nuclide, g/liter	11.6	11.6	7.3
Atomic ratio of hydrogen to fissile nuclide*	2250	2250	3630
Areal density of fissile nuclide, g/cm^2	0.40	0.40	0.25

*Lower limit

4.2.2   General Discussion

Nuclear criticality depends on the volume, mass, and concentration of the fissile materials. Criticality is a great concern when aqueous solutions are involved because water is a very good neutron moderator. Potential conditions of inadvertent criticality may occur during a fire fighting situation where water is introduced. In all cases, the chemical process associated with separation and purification operations require that containers used in the process be in criticality safe configurations. However, accidents initiated by corrosion of equipment, which can lead to leakage, may pose a criticality concern.

Many short-lived fission products are generated when a nuclear excursion occurs. The fissile materials commonly found in commercial spent nuclear fuel include ^{233}U, ^{235}U and ^{239}Pu. Inadvertent criticality may occur with sufficient mass, high fissile-to-moderator ratio, and certain geometrical configuration. Good moderators contain hydrogen, such as water and hydrocarbons. Once criticality occurs, the measure of the fission energy is often referenced by the number of fissions. Typically, a fission produces about 200 MeV (3.204×10^{-11} joules). An excursion of 10^{18} fissions in 1 second would produce 32 MW of thermal power. In addition, the fission products produced by this number of fissions may be significant if the duration of the excursion lasts for a significant amount of time (see Table 4-6 for the duration of the excursions in past criticality accidents). A representative fission value and time can be input into a code such as ORIGEN, to obtain the fission product inventory. For a spent fuel solution criticality accident with a total of 10^{19} fissions, the anticipated quantities and radionuclides generated are shown in Table 4-8. As shown in this table, the quantities of the noble gases and iodine fission products in terms of activity are given from 0 to 8 hours following the excursion.

Table 4-8   Activity of important gaseous and volatile radionuclides released during a 10^{19} fission excursion in spent fuel solution*

Nuclide	Half-Life	Radioactivity, Ci[1]		
		0 to 0.5 hr	0.5 to 8 hr	Total
Kr-83m	1.8 hours	3.7E+00	3.3E+01	3.7E+01
Kr-85m	4.5 years	1.6E+01	1.5E+02	1.7E+02
Kr085	1.7 years	1.5E-04	1.4E-03	1.6E-03
Kr-87	76.3 minutes	1.0E+02	9.0E+02	1.0E+03
Kr-88	2.8 hours	6.5E+01	5.9E+02	6.6E+02
Kr-89	3.2 minutes	4.1E+03	3.7E+04	4.1E+04
Xe-131m	11.9 days	3.8E-04	3.5E-03	3.9E-03
Xe-133m	2.0 days	5.5E-02	1.9E+00	2.2E+00
Xe-133	5.2 days	1.3E+00	2.3E+01	2.7E+01
Xe-135m	15.6 minutes	1.1E+01	2.8E+03	3.3E+03
Xe-135	9.1 hours	1.6E+01	3.5E+02	4.1E+04
Xe-137	3.8 minutes	3.8E+03	4.2E+04	4.9E+04
Xe-138	14.2 minutes	1.2E+03	9.5E+03	1.1E+04
I-129	2x10⁷ years	3.9E-10	4.3E-10	4.3E-10
I-131	8.1 days	1.8E-01	1.6E+00	1.8E+00
I-132	2.3 hours	6.7E-01	6.1E+00	6.7+00
I-133	0.8 hour	3.5E+00	3.1E+01	3.5E+01
I-134	52.6 minutes	4.8E+01	4.3E+02	4.8E+02
I-135	6.6 hours	1.2E+01	1.0E+02	1.2E+02

*Spent fuel contains < 5% U-235 enrichment [DOE 1994]
[1]Total Ci. The assumption of cumulative yield is very conservative (i.e., it does not consider appropriate decay schemes).

4.3 Fire

This section presents an overview of the historical fire accidents at reprocessing facilities and discusses the pertinent phenomena associated with aerosol generation and transport due to fires in Section 4.3.1 and Section 4.3.2, respectively.

4.3.1 History

A fire in a fuel reprocessing facility is possible due to physical heating and/or heating from chemical reactions. Table 4-9 lists fire accidents that have occurred in reprocessing facilities. In most cases, a fire also occurs in conjunction with an explosion.

Table 4-9 A summary of fire incidents at the fuel reprocessing facilities

Facility	Year	Accident	
		Type	**Description**
Hanford [Cadwallader 2005]	1951	Fire – human error	A laboratory room suffered a fire. Apparently, nitric acid on rags used for cleaning and decontaminating parts had been boxed for disposal. The rags spontaneously combusted in the waste storage room of a small laboratory building. The security patrol noted smoke from the ventilation stack after the normal work day and heard crackling noises over the night alarm public address system. Firefighters arrived and used water to extinguish the fire. The fire burned for about four hours.
Hanford [Cadwallader 2005]	1963	Fire – equipment failure	In the PUREX plant, resin in a plutonium extraction column caught fire. The fire spread plutonium contamination throughout the building and some outside of the building.
La Hague [Scheider 2001]	1981	Fire – equipment failure	Uranium metal caught fire following a mechanical shock during operations. The maximum measured level of air contamination, 700 Bq/m³, was reached 10 hours after the beginning of the fire. Graphite elements had been burning for 24 hours in a waste silo.

4.3.2 General Discussion

A fire starts with ignition of a combustible material (solid or liquid) in a room. The combustible material reacts with the available oxygen and releases heat and combustion products (including soot). The heated combustion products rise as a buoyant plume above the fire, entraining air as the plume rises to form a hot gas layer of the combustion products and fresh air under the ceiling. As the hot air rises, cooler air fills in to provide oxygen to sustain the fire. Eventually, this layer grows by pushing air out of any opening that connects with the room. Any gases and combustion products that leave the room containing the fire forms a hot gas layer near the ceiling of a connecting room. Although this room may not have a fire source, a layer grows downward from the ceiling as more gas flows into the room from the fire.

The basic concept and model of a fire in a room and fluid exchange between connecting rooms have been documented [Tanaka 1983, Ho 1988, and Beall 1997].

In terms of modeling an accident involving a fire, the important phenomena include:

1. Heat released from the fire is directly related to the amount of combustibles available to burn and available oxygen in the room. Both energy and duration are important parameters to determine the extent of the accident.

2.   Thermal mass in the room and surrounding rooms are the major heat sinks for the fire. The heat fluxes can be calculated from the fire to the walls, ceiling and floor and other objects in the room. (No credit is usually given for sprinklers.)

3.   Hot gas layer build-up near the ceiling is an important phenomenon in a fire. The stratification behavior captures the thermal plume from the fire. The layer is dynamic in terms of the interaction with the plume, incoming cooler flow from doors and ventilation system (mixing). As the hot gas layer grows, the temperature of the room fire increases, which may initiate or induce a secondary accident such as another fire and/or explosion.

4.   Combustion products are important because they influence the transport of radionuclides from processing solution to other locations within the facility.

4.4   Spills and Leaks

This section presents an overview of historical spill accidents at reprocessing facilities and discusses the pertinent phenomena associated with aerosol generation and transport due to spills in Section 4.4.1 and Section 4.4.2, respectively. This section, similar to the overall report, specifically focuses on potential events that lead to creation of vapors and aerosols that could be released into the atmosphere. Spills and leaks can lead to groundwater contamination as well, but that topic is not discussed here.

4.4.1   History

Spill accidents in a fuel reprocessing facility are mainly caused by equipment, piping, or container failure. The failures are often caused by a combination of corrosion and pressure transients. Corrosion results from the reaction of strong acid solutions with vessel and piping components. The primary cause of pressurization is chemical reactions inside the process vessel or holding tank. As given in Table 4-10, eleven spill accidents have been documented.

Table 4-10   Spill accidents in fuel reprocessing plants

Facility	Year	Accident	
		Type	**Description**
Hanford [Cadwallader 2005]	1953	Pressurized spill – operator error	During a process evolution, an acid solution and a caustic solution were improperly mixed. Metal waste supernatant was being pumped from a holding tank to a blend tank to prepare process feed for the TBP. Approximately 2,800 gallons of 60% nitric acid was first added to the blending tank. The metal waste addition began, and the agitator for the tank was started. After about 100 minutes, a supervisor noted yellowish ooze coming from the tank cover blocks. As he rushed to the controls to stop the supernatant flow, a geyser of liquid quickly rose about 10 meters in the air for about 30 seconds, and the prevailing wind carried the geyser liquid onto the supervisor and other nearby workers. They were contaminated at ~4,000 counts per minute (cpm) and they had stinging sensations from the liquid. The supervisor called to shut the roadway that had been contaminated by the geyser. Investigation revealed that the operator had activated the agitator switch but the agitator had "bumped;" it started to operate but did not continued to run. The operator had not paused to verify that the agitator was continuing to run. Trough areas near the blend tank contained up to about 6-inch deep yellowish liquid that read a 35 rem/hour at a distance of 6-inches.
Hanford [Cadwallader 2005]	1956	Spill – equipment failure	At the REDOX plant, two process solution (highly concentrated acidic plutonium product) spills were found on the floor behind the control panel. The solution entered the control room by an instrument air line. The instrument airline operated a flow control valve in the process transfer line from the final product concentrator tank to a receiver vessel. When air pressure was relieved from the flow control valve, the air was intended to bleed harmlessly into the control room. The flow control valve stem had a bellows to protect it, but the bellows had failed and process solution had, over time, collected in the valve superstructure. The bleed hole in the valve superstructure still had the manufacturer's dust plug in place. Over time, with no other pathway, the process solution backed up into the air line and out of the vent in the control room. The operator who noted the survey meter "break down" was contaminated on all skin surfaces to over 667 Bq. Nasal smears read 500 Bq and all of his clothing was contaminated. A rough estimate was that only about 350 cm^3 was spilled in the control room, and the clean-up cost was $2,000 - $2,500.
Savannah River Site [Cadwallader 2005]	1960	Spill – equipment failure	A valve corridor was highly contaminated by leaking coolant water. The coolant water had become contaminated by high activity waste from the PUREX process, and then the water had leaked through a defective waste evaporator reboiler. Approximately 5,000 Curies were released with the water. Most of the released radioactivity was contained in the building and was flushed to the waste handling facilities. Minor amounts of liquid were released to the seepage basins outside the building. A series of operating errors and miscommunications allowed the initial leakage water to flow back through open valves on steam traps and then out into the corridor. The corridor was flooded with water, and the contaminated water flowed down a stairwell and spread widely throughout normally nonradioactive sections of the building. Radiation readings as high as 400 rad/hour at 30 cm were measured after the incident.

Table 4-10   Spill accidents in fuel reprocessing plants

Facility	Year	Accident	
		Type	**Description**
Hanford [Cadwallader 2005]	1961	Spill – equipment failure	At Hanford Works in the PUREX plant, the acid waste concentrator used a heat exchanger to help condense acid. The heat exchanger had to be taken out of service after only 18 months of operation. The failure was caused by improperly placed weld bead at several tube to tube sheet welds. The welder had laid bead by hand and apparently had deviated by as much as 50 mils (1.27 mm). The tube wall thickness was affected by the weld placement and this led to premature failure of the tube bundle. The investigators also noted that some tubes had been plugged with solid material and other tubes had solid films, which would have reduced the effective heat transfer.
Savannah River Site [DOE/EH-0555]	1972	Pressurized spill – chemical reaction	A pressure surge in a waste evaporator being brought up to operating temperature caused the eructation of approximately 6,000 pounds of process solutions from the vessel. The solution was a dilute mixer-settler flush of 1 M nitric acid and 0.1 M HAN at the time of accident.
Savannah River Site [DOE/EH-0555]	1978	Pressurized spill – chemical reaction	A sudden eructation occurred from a tank thought to be empty after addition of 25% (~2,000 pounds) nitric acid. However, the tank had 5% HAS in the heel of the tank. The eructation sprayed 3 individuals.
Savannah River Site [DOE/EH-0555]	1980	Pressurized spill – chemical reaction	Dilute HAS/nitric acid solution in a tank was inadvertently heated over a period of a few days due to a leaky steam coil. The heating allowed the autocatalytic condition to be reached, causing the release of acid solution to the surrounding of the tank. A ruptured elbow, bent piping and ejection of the anchor bolts are the result of this accident.
Hanford [DOE/EH-0555]	1989	Pressurized spill – chemical reaction	An exothermic reaction involving a solution of HAN, nitric acid and hydrazine occurred in a one-inch chemical makeup line in the PUREX plant aqueous makeup area. The chemical reaction pressurized the isolated piping and blew out a gasket at the flange connection.
Savannah River Site [DOE/EH-0555]	1996	Pressurized spill – chemical reaction	The temperature of a tank containing HAN in strong nitric acid was not monitored and rose to 40 ˚C where an autocatalytic event resulted in an eructation of 250 gallons.
THORP [DOE2005-11]	1998	Spill – equipment failure	Erosion of an outlet pipe in the dissolver cell resulted in the leak of highly radioactive dissolver solution into the secondary containment. Despite sump-level indication, sampling and contaminated radiological probes, this leak went unnoticed for years.
THORP [DOE2005-11], [Todd 2008]	2005	Spill – equipment failure	A pipe failure resulted in about 83,000 liters of highly radioactive dissolver solution leaking into the stainless-steel lined feed clarification of the THORP facility in 2005. This solution contained about 19 metric ton of uranium and plutonium. The leak went undetected for months (possibly since July 2004), before being discovered.

Since complex chemical processes are used in highly radioactive spent fuel reprocessing, the chemical and radiation conditions are such that corrosion of equipment is a major concern in a spill accident. The corrosion problems could occur in (1) dissolvers where hot or boiling nitric acid is used to dissolve the spent fuel; (2) solvent extraction vessels for the separation processes of uranium, plutonium and fission products; (3) evaporators to concentrate these materials in hot nitric acid; and (4) storage tanks for radioactive waste of warm concentrated nitric acid [Mudali 1993]. To prevent and mitigate these types of accidents, frequent visual inspection of the process equipment is necessary.

4.4.2   General Discussion

Spill accidents are the least energetic event compared with inadvertent criticality events, fires, and explosions. Often a spill may not be detected immediately. Impaction of liquid against a solid surface may create aerosols and air flow may entrain some of the material in a pool created by the spill. Many aerosol models for spill accidents focus on air flow parallel to the solution pool to determine the amount entrained from the pool surface. A more energetic spill is based on a free fall concept from a few feet to 10 feet. Break-up phenomena account for the aerosol generation/suspension when the liquid hits the ground.

5.   ACCIDENT AND AEROSOL GENERATION MODELING

In previous sections of this report, chemical separation processes and anticipated accidents (based on historical experience) are described to provide the context for identifying models and methodologies needed to characterize a source term. Characterization of a source term requires understanding the evolution of hazardous materials from the initial accidental release, through transport within the facility, and finally release to the environment.

To properly characterize a source term, a sequential process is presented in this chapter. First, a determination of the available inventory and form of the material associated with a process or system within a reprocessing facility is discussed in Section 5.1. Then the more likely accidents are discussed in Section 5.2. For each accident the current techniques for estimating the released aerosol are given. Current techniques are primarily empirical and therefore limited, and may not be appropriate for reprocessing facilities. Therefore, Section 5.3 describes a basic principles approach that is proposed to replace the empirical approach for some phenomena. The transport of material through the facility is not discussed.

5.1   Inventory in Operations

This section describes the spent fuel inventory during each step of the operations in a typical fuel reprocessing facility. This inventory greatly depends on the initial spent fuel received at the facility. From the operational history and cool down period of a given fuel, a determination of the inventory can be made to characterize the composition of the fuel, i.e., fission products and transuranics present. Fuel characterization is readily performed using ORIGEN or a similar code. As an example, the spent fuel presented has a burnup of 60 GW-day per metric ton of fuel with a minimum cooling period of 5 years. Table 5-1 shows the spent fuel inventory in terms of elements per metric ton for a typical low-enriched, light-water-reactor spent fuel for two burnups, 45 and 60 GWd/t, with 5- and 30-year cooling periods. The burnup in commercial plants could be lower than those used in this report. Both mass and decay heat power are presented in this table. The decay heat power comes from the isotopes of the elements reported.

Table 5-1   **Typical low-enriched light water reactor spent fuel inventory (per metric ton of fuel) for two cooling periods and burnup ranges [NEA 2012-15]**

Component	5 years, Cooling time				30 years, Cooling time			
	Burn-up, 45 GWd/t Spent fuel		Burn-up, 60 GWd/t Spent fuel		Burn-up, 45 GWd/t Spent fuel		Burn-up, 60 GWd/t Spent fuel	
	kg/t	W/t	kg/t	W/t	kg/t	W/t	kg/t	W/t
Zr	362		362		362		362	
SS	57		57		57		57	
Inconel	21		21		21		21	
Total Clad	440		440		440		440	
Xe	7.12		9.59		7.12		9.59	
Kr	0.50	14	0.63	17.4	0.48	2.9	0.61	3.5
H-3	0.00008		0.00010		0.00002		0.00002	
C-14	0.00013		0.00019		0.00013		0.00019	
I	0.26		0.36		0.26		0.36	
Cs	3.69	493	4.76	762	3.02	79	3.87	105
Ba	2.23	474	3.05	625	2.96	266	3.94	351
Sr	1.11	106	1.41	130	0.87	57	1.03	70
Y	0.64	508	0.8	621	0.64	274	0.8	335
Zr	4.82		6.26		4.82		6.26	
Sb	0.021	11.6	0.028	15	0.017	0	0.024	0
Mo	4.60		6.06		4.60		6.06	
Tc	1.07		1.36		1.07		1.36	
Ru	2.96	1.2	4.16	1.6	2.96	0	4.15	0
Rh	0.60	190	0.73	251	0.60	0	0.73	0
Pd	1.68		2.68		1.68		2.69	
Ag	0.09		0.14	0.6	0.09		0.14	
Total NM	11.0	191	15.1	253	11.0	0	15.1	0
Gd	0.15		0.31		0.18		0.346	
Eu	0.19	60	0.26	90	0.17	7.9	0.23	11.9
Sm	1.06		1.37		1.12		1.43	
Pm	0.063	21	0.062	21	0	0	0.84	0
Total HREs	1.463	81	2.002	111	1.47	7.9	2.846	11.9
Ce	3.21	10	4.23	10	3.21	0	4.22	0
Pr	1.54	114	2.01	113	1.54	0	2.01	0
Nd	5.57		7.31		5.57		7.31	
La	1.67		2.19		1.67		2.19	
Total LREs	12.0	124	15.7	123	12.0	0	15.7	0
Total RE	13.5	205	17.7	234	13.5	8	18.6	12
Other FP	31.4	1786	42.0	2409	31.2	679	41.6	865
U	941	0.06	923	0.06	941	0.06	923	0.06
Pu	11.2	164	12.6	283	10.2	138	11.5	236
Np	0.57	0.01	0.78	0.02	0.57	0.01	0.78	0.02
Am	0.51	47	0.74	58	1.38	146	1.78	178
Cm	0.033	88	0.113	292	0.014	34	0.0497	112
Total TRUs	12.3	299	14.2	633	12.2	318	14.1	526

*NM: Noble Material, RE : Rare Earth, LRE : Light Rare Earth, HRE : Heavy Rare Earth

A representative list of the fission products that contribute to the decay heat as a function of the cooling time in terms of years is shown in Figure 5-1[3]. As shown in this figure, the principal fission products at times < 10 years are ^{134}Cs, ^{137}Cs + ^{137m}Ba progeny, ^{90}Sr + ^{90}Y progeny, ^{106}Rh, ^{154}Eu, ^{144}Ce + ^{144}Pr progeny and ^{147}Pm. Based on a DOE report [DOE/SNF078], elemental decay heat as shown in Table 5-1 is primarily from Kr as ^{85}Kr, Ba as ^{137m}Ba, Sr as ^{90}Sr, Y as ^{90}Y, Sb as ^{125}Sb, Ru as ^{106}Ru, Rh as ^{106}Rh, Eu as ^{154}Eu, Pm as ^{147}Pm, Ce as ^{144}Ce, Pr as ^{144}Pr, U as ^{234}U and ^{238}U, Pu as ^{238}Pu, ^{239}Pu and ^{240}Pu, Np as ^{137}Np, Am as ^{241}Am, and Cm as ^{242}Cm.
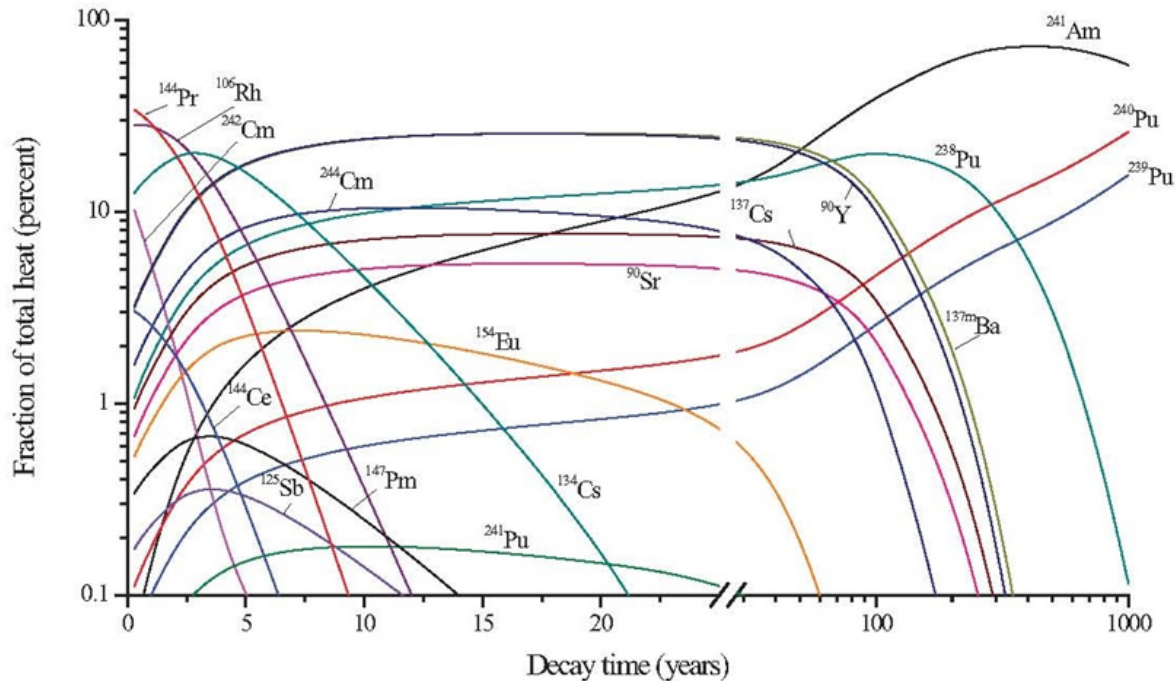
Figure 5-1   Important fission products, which contributed to decay heat for typical light water reactor fuel for cooling times from 1 to 1,000 years [OECD 2011]

The accident type and process stage determines the material suspended as vapors and aerosols. For example, spent fuel before the dissolution process is in solid form, and may be contained in a fuel rod. Spent fuel in the extraction and dissolution processes is in solution, and therefore behaves differently. This, understandably, requires resolution of materials into the isotopic amounts and physical forms throughout the various stages of the reprocessing facility.

An example of the inventory during the operations of a UREX type of extraction is shown in Figure 5-2 and Figure 5-3 for 5-year and 30-year cooled fuel, respectively. As shown in these figures, most of the volatile fission products are released during the voloxidition process. In this process, UO_2 solution is heated to convert UO_2 to U_3O_8, and volatiles escape as in the proposed AREVA plant [Bader 2011]). During the dissolution operation, the off-gas contains iodine vapors that are captured by a silver impregnated zeolite bed (it can be found in the proposed AREVA plant [Bader 2011]). In this dissolution operation hulls and other assembly hardware waste are produced (it can be found in the proposed AREVA plant [Bader 2011]).

---

[3] This figure is based on calculations performed for an enrichment of 4.5 wt% and 50 GWD/T burnup.

The remaining solution operations, after the dissolution operation, separate out U, U-Pu-Np, and various fission product streams (it can be found in the proposed AREVA plant [Bader 2011]). Similar determination of fission product and transuranic inventories need to be established to quantify the material at risk in the event of an accident.
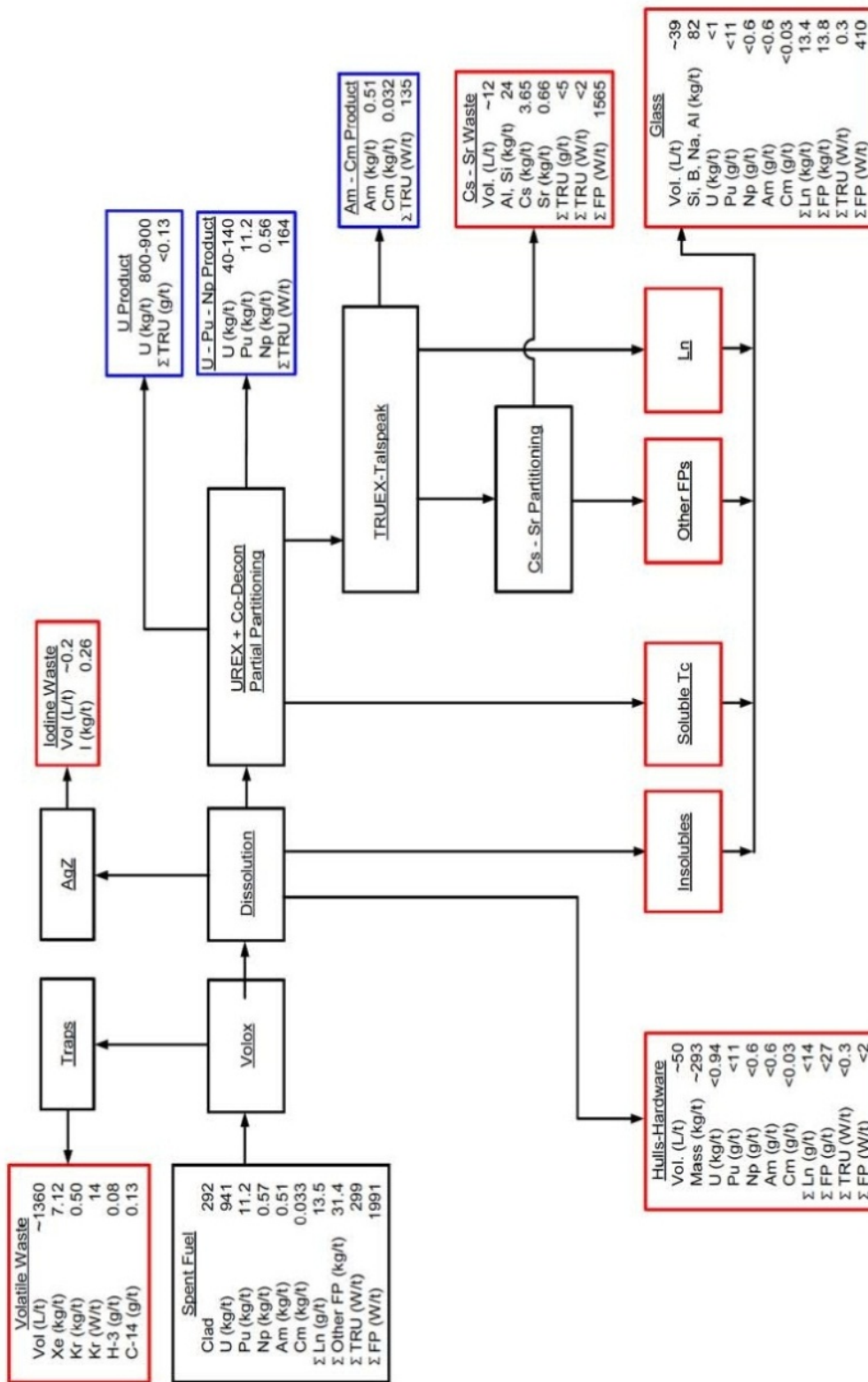
Figure 5-2   Operation inventory for an UREX type extraction process (with no Pu separated out) of a 5-year cooled and 45 GWd/t burnup [NEA 2012-15].

U Product
| U (kg/t) | 800-900 |
| Σ TRU (g/t) | <0.13 |

U – Pu – Np Product
U (kg/t)	40-140
Pu (kg/t)	10.2
Np (kg/t)	0.56
Σ TRU (W/t)	138

Am – Cm Product
Am (kg/t)	1.38
Cm (kg/t)	0.01
Σ TRU (W/t)	180

Cs – Sr Waste
Vol. (L/t)	~11
Al, Si (kg/t)	26
Cs (kg/t)	2.99
Sr (kg/t)	0.86
Σ TRU (g/t)	<5
Σ TRU (W/t)	<2
Σ FP (W/t)	669

Glass
Vol. (L/t)	~40
Si, B, Na, Al (kg/t)	84
U (kg/t)	<1
Pu (g/t)	<11
Np (g/t)	<0.6
Am (g/t)	<0.6
Cm (g/t)	<0.03
Σ Ln (kg/t)	13.4
Σ FP (kg/t)	14.5
Σ TRU (W/t)	0.2
Σ FP (W/t)	15

Iodine Waste
| Vol (L/t) | ~0.2 |
| I (kg/t) | 0.26 |

Volatile Waste
Vol (L/t)	~1360
Xe (kg/t)	7.11
Kr (kg/t)	0.48
Kr (W/t)	2.9
H-3 (g/t)	0.02
C-14 (g/t)	0.13

Spent Fuel
Clad	292
U (kg/t)	941
Pu (kg/t)	10.2
Np (kg/t)	0.57
Am (kg/t)	1.38
Cm (kg/t)	0.014
Σ Ln (g/t)	13.5
Σ Other FP (kg/t)	31.2
Σ TRU (W/t)	318
Σ FP (W/t)	679

Hulls-Hardware
Vol. (L/t)	~50
Mass (kg/t)	~293
U (kg/t)	<0.94
Pu (g/t)	<11
Np (g/t)	<0.6
Am (g/t)	<0.6
Cm (g/t)	<0.03
Σ Ln (g/t)	<14
Σ FP (g/t)	<27
Σ TRU (W/t)	<0.3
Σ FP (W/t)	<2

Process units: AqZ, Traps, Volox, Dissolution, UREX + Co-Decon Partial Partitioning, TRUEX-Talspeak, Cs - Sr Partitioning, Ln, Other FPs, Soluble Tc, Insolubles

Figure 5-3 Operation inventory for a UREX Type extraction process (without Pu separated) of 30-year cooled fuel with 45 GWd/t burnup [NEA 2012-15].

5.2   Source Term Potentials and Release Mechanisms

Currently, many non-nuclear-reactor facilities, such as those at many Department of Energy sites, use a five-factor formula to identify their source term releases [DOE 1994]. These factors are all multiplied to arrive at a source term. Each of the factors is defined below:

1.   MAR – for our purposes, material-at-risk is the radioactive inventory of the spent fuel in an operation. For a criticality accident, additional radioactive inventory as a result of fissions are included in the computation of MAR. For a tank containing spent fuel solution materials, the radioactive inventory in the tank is the MAR.

2.   DR – damage ratio is the fraction of MAR involved in the accident. For example, if a tank bursts, the fraction of the initial MAR that is spilled is the DR.

3.   ARF – airborne release fraction is the fraction of MAR that is airborne.

4.   RF – respirable fraction is the fraction of MAR that is respirable. For example, the fraction of the airborne spill content that is in the respirable size range. Particles smaller than about 20 μm in aerodynamic diameter are considered respirable.

5.   LPF – leak path factor is the fraction of MAR released out of the facility. For example, the fraction of respirable aerosol from the airborne spill content that gets out of the facility. This factor considers the room from which the accident occurred to the various open pathways to release out of the facility.

Based on the discussions in Section 4, four major accident categories can be identified: criticality, explosion, fire, and spill. Among these accident categories, explosions contain the most energy and may fail the confinement boundary of the facility especially in locations where direct radiation levels are low and robust shielded structures are not required. However, a smaller explosion may only damage the primary container of the MAR, and possibly the room in which the container resides. In this case, a LPF calculation is necessary to determine source term from the facility. Therefore, the following sections provide more detail about the phenomena associated with criticality, explosion, fire and spill accidents, including any past accidents in fuel reprocessing facilities.

Several computer codes are widely available to determine the source term from a facility. Many of these codes came from the severe accident analysis of nuclear power reactors. Below is a list of the system codes which can be used for source term analysis:

1.   MELCOR – This code was developed for the Nuclear Regulatory Commission (NRC) by Sandia National Laboratories. Since its inception in 1980s, MELCOR has been used in analyzing nuclear power reactor accidents, including spent fuel pool accidents. In addition, MELCOR is used for non-reactor applications to estimate the source term from nuclear and non-nuclear facilities at Department of Energy (DOE) sites. These facilities include:
        a.   Plutonium processing facilities
        b.   Weapon assembly facilities
        c.   Weapon material processing facilities
        d.   Nuclear material processing facilities
        e.   Nuclear material testing facilities

2. MAAP – This code was developed by Fauske and Associates for the U.S. nuclear utilities. The code is used for supporting licensing of commercial nuclear power plants.

3. FATE™ – This computer code was developed by Fauske and Associates and is intended for fuel cycle and non-reactor facilities. It has been used for Hanford waste facilities [NS-ES-0194].

4. ASTEC – This code was developed by IRSN (Institut de Radioprotection et de Surete Nucleaire) and GRS (Gesellschaft fur Anlagen und ReaktorSicherheit), and serves a similar function as MELCOR [Lajitha 2008].

The above system computer codes can predict the room-to-room transport based on the pathways in the facility during an accident. Almost all of these system codes have been developed for nuclear reactor applications. Only the FATE™ code includes specific models for fuel cycle facilities and other facilities [NS-ES-0194]. MELCOR has been used for estimating the LPF values for DOE nuclear and non-nuclear facilities as indicated in Item 1 above. MELCOR can also be used to model spent fuel pools. To model a spent nuclear fuel reprocessing facility, additional models need to be added to the existing system codes to simulate accidents in a reprocessing facility. The next subsection discusses the phenomena threat need to be modeled in the system codes, and the data or correlations that are available.

5.2.1 Explosions

Explosions can occur in the fuel reprocessing facilities when process vessels over pressurize as a result of chemical reactions or physical processes. Physical explosions are a result of process vessels not being able to relieve pressure because of a cooling system or pressure relief valve failure. Chemical explosions include "red oil" reactions, HAN reactions, hydrogen combustion, and combustion of other gases or solids (such as zirconium fines) inside process vessels. Of course, the impacts of these explosions are not restricted to vessels and containers containing the unstable reactants, but may also affect other process equipment, confinement barriers, or ventilation system. The potential for a steam explosion is possible in the waste vitrification facility. However, this section primarily discusses explosions associated with process vessels as a result of chemical reactions.

An explosion can be delineated into various phases, each requiring predictive modeling to determine the potential aerosol and vapor generation that is available for release. The following subsections include methods for determining vessel failure pressure, airborne release, evaporation, and overpressure blast effects.

5.2.1.1 Vessel Failure Pressure

When an explosion occurs inside process equipment, it must first fail the vessel to be of significance. An explosion that leads to vessel failure is called a pressure vessel burst (PVB) [AIChE 2010]. Examples of PVBs include (a) chemical reaction or combustion inside the vessel, (b) overpressure due to external sources, such as high pressure supply from pressurized equipment (pumps), and (c) failure of the pressure relief valves if overpressure of the vessel is an anticipated situation. The equations presented below can be used to estimate the bursting pressure of a process vessel [NUREG/CR-4593].

The hoop-stress equation is commonly used to determine the limiting stress on thin-walled process vessels

$$\sigma_t = S_p = \frac{P\, d_o}{2\, t}$$

Equation 5.1

where σ_t= ultimate tensile strength (psi), S_p= hoop stress (psi), P= burst pressure (psi), d_o= outside diameter of vessel (inches), and t= thickness of the vessel (inches). To solve for P, Equation 5.1 becomes

$$P = \frac{2\, t\, \sigma_t}{d_o}$$

Equation 5.2

An empirical correlation for P that accounts for the fatigue of the process vessel due to repeated cycling of pressure or by the notching of the vessel walls is given by the Bach equation [NUREG/CR-4593]:

$$P = \frac{\sigma_t\left(d_o^2 - d_i^2\right)}{1.3\, d_o^2 + 0.4\, d_i^2}$$

Equation 5.3

where d_i = inside diameter of vessel (inches).

A modified Svensson equation for P is given by:

$$P = \sigma_t F_{cyl}\left(\frac{2\, t}{d_o}\right)\left(1 - \frac{t}{d_i}\right)$$

Equation 5.4

where F_{cyl}= cohesion factor for cylinders and is given as:

$$F_{cyl} = \left(\frac{0.25}{\varepsilon_u + 0.227}\right)\left(\frac{e}{\varepsilon_u}\right)^{\varepsilon_u}$$

Equation 5.5

where e= base of natural log, and ε_u= true strain rate at maximum log.

To demonstrate the differences in computing the burst-pressure from Equation 5.2 to Equation 5.4, consider a 304 stainless steel process vessel with a 48-inch outer diameter and 0.25 inch thickness, and the material properties given in Table 5-2. The burst pressures calculated with Equation 5.2, Equation 5.3, and Equation 5.4 are 854, 1005, and 643 psi, respectively.

In addition to empirical correlations, as discussed above, computational tools are available to simulate in detail the rupturing of a vessel of essentially arbitrary geometry, materials of construction, explosive energy, and explosive location within the vessel. The simulation provides not only the rupture size, geometry, and location, but also can estimate the mass and particle size distribution of liquid ejected. The simulation provides the liquid mass that (1) remains in the vessel, (2) settles to the ground or adheres to surfaces, and (3) becomes aerosol. These results are obtained by using a structural mechanics code to solve the solid mechanics problem of material deformation on short time scales, as discussed in Section 5.2.

Table 5-2 Process vessel characteristics and its material properties [NUREG/CR-4593]

Facility Type	Process Vessel Materials	Vessel Thickness (inches)	Outside Diameter (inches)	Length (inches)
MOX fuel fabrication	Stainless steel	Up to 0.25	3.6 – 72	13.8 - 384
Fuel reprocessing	Stainless steel Titanium	Up to 0.25 --	3.6 – 132 72	216 120
Solidification*	Stainless steel Hastelloy X	Up to 0.25 --	-- 36	-- 120
Material	**Ultimate Tensile Strength (psi)**			**True Stain Rate, ε_u**
Carbon steel AISI-SAE 1020	65,000			--
304 stainless steel	82,000			0.585
Titanium alloy	144,000			--
Hastelloy X	110,000			--

*Solidification includes a number of methods, including using inorganic additives, organic additives and vitrification

5.2.1.2 Airborne Release Estimate

Extrapolation of experiments used in studies of explosive-induced dispersal of both solids and liquids in a closed chamber of sufficient volume to preclude build-up of substantial pressure were suggested by Steindler and Seefeldt [Steindler 1980]. The experiments are for spherical explosives or cylindrical explosives with a diameter to height ratio of one. Based on the extrapolation, an empirical procedure has been created for producing an airborne release estimate for use in facilities [Steindler 1980]. The steps in this approach are as follows.

1.   Estimate the mass ratio (r_m) as the ratio of the weight of the inert material to the weight of explosive. The explosive used is the TNT-equivalent weight of the explosive material (such as the unstable solvent in the case of the red oil). Table 5-3 shows the TNT equivalence for various systems of combustible/pressurized gases and liquids.

Table 5-3 TNT equivalence for various gas, combustible gas and combustible liquids released during vessel rupture events [NUREG/CR-4593]

Material and Conditions	Pound of TNT Equivalence* Per ft^3 of Material Released under Stated Condition
Gas at 100 psi and 20 ˚C	0.02
Gas at 1000 psi and 20 ˚C	0.42
Gas at 10,000 psi and 20 ˚C	6.53
Argon at 20,000 psi and 20 ˚C	~1.0
Hydrogen gas at 20,000 psi and 20 ˚C	~1.7
Hydrogen explosion at 50% efficiency, including hydrogen gas released to atmosphere at 20,000 psi and 20 ˚C	~170
Mineral oil at 20,000 psi and 20 ˚C	~0.06
Red oil explosion	0.23

*1 pound of TNT is equivalent to the release of 1830 BTU (~5x10^5 cal)

2.   Estimate the mass median diameter (d_g) from Figure 5-4. Note the experimental data only contains data up to r_m of about 15 μm. Anything above this r_m is considered an extrapolation.
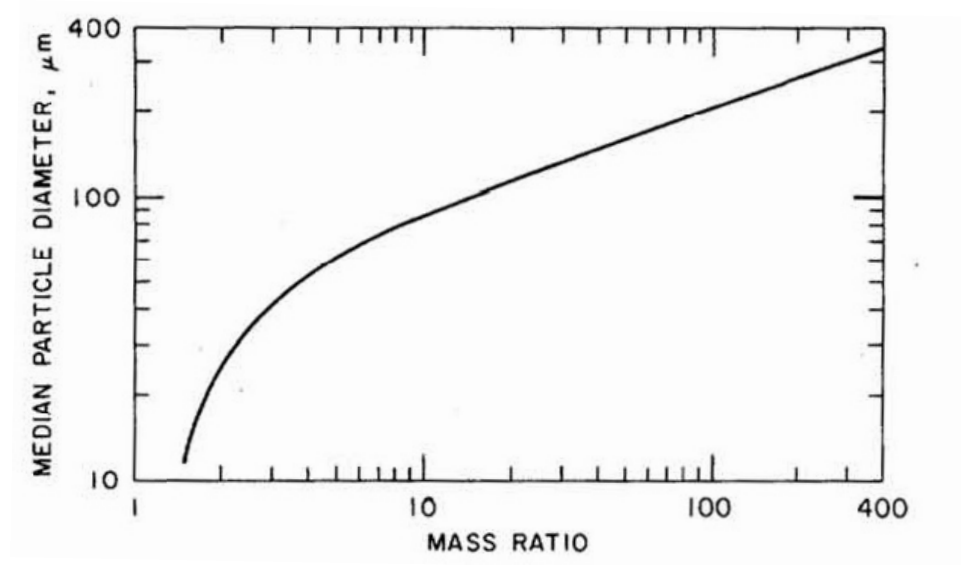
Figure 5-4   Median particle diameter as a function of mass ratio (r_m) [Steindler 1980]

3.   Estimate the ratio of mass airborne per mass of explosive, AR (g aerosol/g explosive) using the expression [NUREG/CR-4593]:

$$AR = 2.783 \, r_m{}^{0.3617}$$

Equation 5.6

or using Figure 5-5.

Note that the airborne release is assumed to be instantly distributed over the room volume, following which settling immediately begins according to Stokes Law, as well as other forms of deposition. Also, an inherent assumption in the transformation from the experimental results to fuel cycle facility applications is the assumption of lognormal distributions with a geometric standard deviation (GSD_g) of 2.
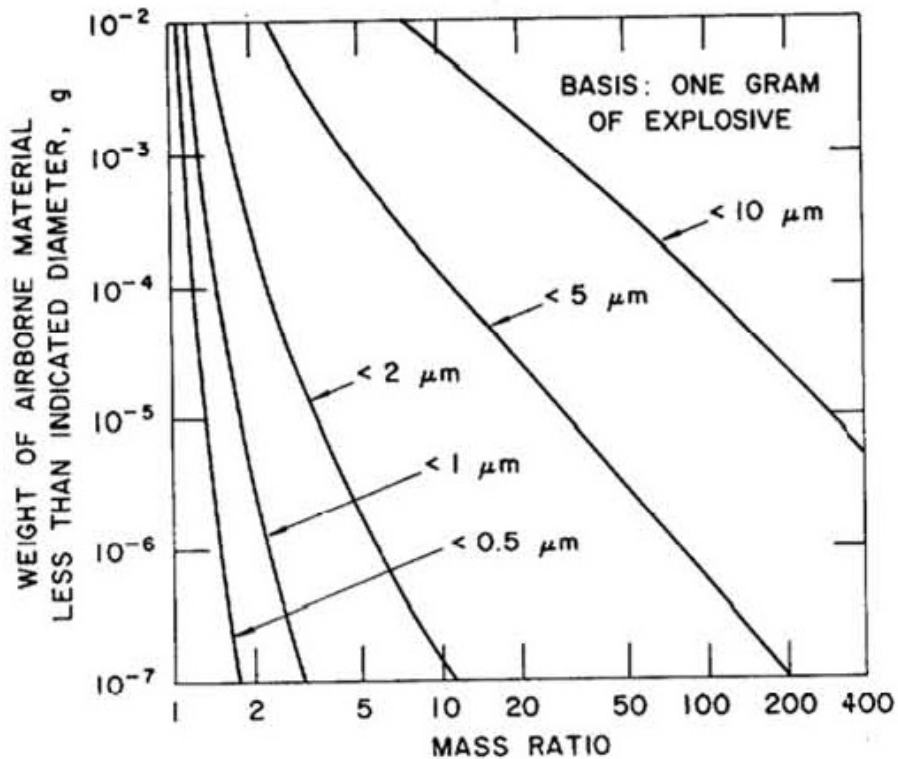
Figure 5-5   Initial AR of airborne versus mass ratio [Steindler 1980]

Additional steps for the procedure have been proposed in [NUREG/CR-4593]. These steps allow for the creation of the particle size distribution. For each size range selected starting from the smallest diameter, it is necessary to calculate the fraction of aerosol in the size range by subtracting the two endpoint values of the cumulative normal frequency distribution, P(Z) (see Figure 3-4 [from NUREG/CR-3593]), where Z is computed as $(\ln d - \ln d_g)/\ln GSD_g$. Finally, the fraction of the aerosol in each range is multiplied by the initial AR computed in Equation 5.6 to obtain the initial aerosol mass for each size range.

Instead of the additional steps described in [NUREG/CR-4593], [Bloom 1994] introduced the fraction of airborne, $AR_{d_1-d_2}$ within the aerosol size range, d_1 to d_2 :

$$AR_{d_1-d_2} = 0.5\,AR\left(erfc\left(\frac{\ln\left(\frac{d_1}{m}\right)}{\sqrt{2}\ln(GSD)}\right) - erfc\left(\frac{\ln\left(\frac{d_2}{m}\right)}{\sqrt{2}\ln(GSD)}\right)\right) \qquad \text{Equation 5.7}$$

Where AR = Equation 5.6, d_i= the aerosol diameter ($d_2 > d_1$) (µm), m = mass median diameter (µm), which is defined as $38.962\,r_m^{0.3617}$, GSD = geometric standard deviation, and $erfc$ = the complementary error function.

Note that [Bloom 1994] had pointed out that the method provided by Steindler and Seefeldt [Steindler 1980] was a "straw man" to encourage further work in this field, but apparently, no further work has been done. Cautions are that the use of the method from Steindler and Seefeldt [Steindler 1980] may require benchmarks from high-explosive experimental data. Red

oil explosions in a pressure vessel may not be similar in terms of energy density, time-scale, etc., to high-explosive experiments used in the model. When a process vessel is ruptured due to an internal explosion and the rupture pressure is less than 500 psi, the airborne release described in Section 5.2.4 may be used.

In recent years, researchers have begun to study the physics of generating liquid droplets by explosions below the liquid surface [Kudryashova 2011, Davydov 2003, Kerdrinskii 1993]. [Kerdrinskii 1993] describes the transformation of a cavitating liquid into a gas-droplet system. The fragmentation of liquids under explosive loading consists of the following four steps:

1. formation and dynamics of bubble clusters in the liquid,
2. unbounded multiplication of cavitation nuclei leading to a foamy structure,
3. breakup of the foamy structure into cavitating fragments, and
4. transition into the droplet state and its evolution.

5.2.1.3 Vaporization

The droplet size distribution from an explosive event changes due to liquid evaporation and the vaporization rate is affected by soluble impurities in the solution. A recent paper [Ishmatov 2010] describes this phenomenon as discussed in the following paragraphs.

In the absence of foreign impurities, liquid evaporation is limited by diffusive transport of vapor away from the drop. For diffusion-limited evaporation the time rate of change of the droplet radius r_d, is given as:

$$\frac{dr_d}{dt} = \frac{D}{r_d \rho_d} \frac{\rho_v - \rho_{air}}{M_w} \qquad \text{Equation 5.8}$$

where D = diffusivity of the vapor, ρ_d = liquid density, ρ_v = vapor density just outside the liquid drop, ρ_{air} = vapor density in air far from the drop and M_w = molecule mass of the liquid.

The effect of soluble impurities is to reduce the vapor density above the liquid. Equation 5.8 becomes

$$\frac{dr_d}{dt} = \frac{D}{r_d \rho_d} \frac{x_d(t) \cdot \rho_v - \rho_{air}}{M_w} \qquad \text{Equation 5.9}$$

where, $x_d(t)$ = mole fraction of liquid as a function of time, which is given by:

$$x_d(t) = \frac{\dfrac{1}{M_{H_2O}(1 - C_i(t))}}{\dfrac{C_i(t)}{M_i} + \dfrac{1 - C_i(t)}{M_{H_2O}}} \qquad \text{Equation 5.10}$$

where M_{H_2O} = water molar mass, $C_i(t)$ = impurity concentration in solution as a function of time and M_i = impurity molar mass.

In Table 5-4 the above equation is used to calculate the evaporation time of water from 20% salt solution drops. As shown in the table, small drops evaporate faster for the same reduction in size.

[Ishmatov 2010] applies the above analysis to estimate the initial aerosol distribution when the aerosol is a result of pulsed explosive spraying of the liquid as shown in Figure 5-6. As shown in this figure, the explosive is located at the bottom of the apparatus and the liquid splashes on the reflector to form the aerosol. Figure 5-7 shows the final aerosol size distribution measured in the experiment (a), and the extrapolated initial size distribution using the above equations (b). The authors of this paper indicate that this pulsed method would yield high velocity droplets up to 300 m/s resulting in high flow turbulence. The evaporation of liquid aerosol is a complex process. Unfortunately, not enough detail is given in the paper to determine the conditions of the experiments or the measurement technique.

Table 5-4   Time of complete evaporation of water from aerosol drops of a 20% salt solution [Ishmatov 2010]

Initial drop size, μm	100	50	40	30	20	15	10	5
Final size, μm	45	22	18	13	9	7	4	2
Evaporation time, s	17	3.85	2.44	1.32	0.68	0.42	0.20	0.04
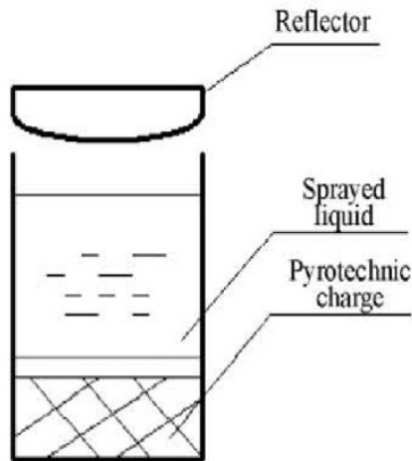
Figure 5-6   Explosive-induced liquid aerosol experiment [Ishmatov 2010]
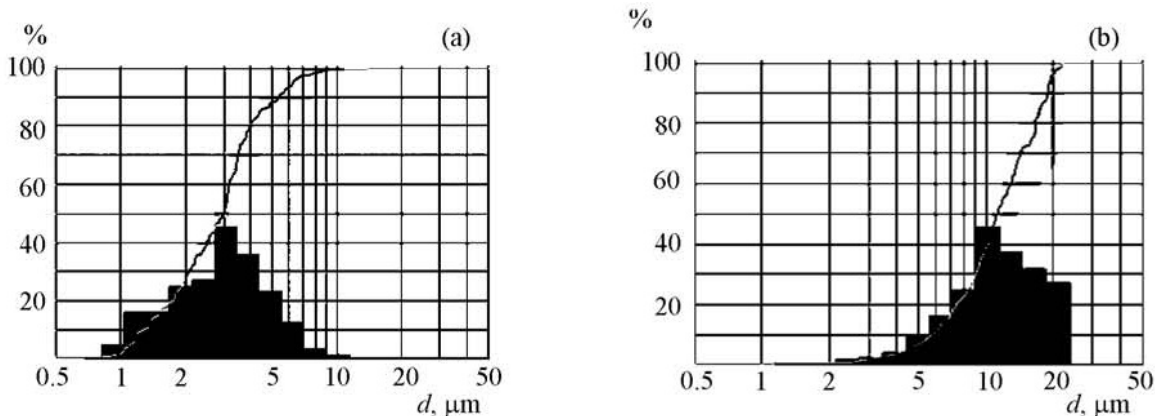
Figure 5-7   Aerosol size distribution for (a) final aerosol distribution in measurement (b) initial aerosol distribution calculated [Ishatov 2010]

5.2.1.4   Blast Models

There are several simplified empirical models available to predict the destruction of walls and ceiling within a room where an explosion has occurred. In this section, two models are discussed:

- Venting model
- Overpressure model

<u>Venting Model</u>

Using the methodology discussed in [Rodriguez 1995], the compartment pressure can be estimated. Furthermore, one can assume that the same thermodynamic relation (Equation 5.11) can be extended to estimate the pressures of the secondary compartments which are adjacent to the explosion compartment.   This method may break down if the explosion is a detonation instead of deflagration. Also, the thermodynamic relation may not be correct if the explosion is directional and the expanded gas jet released from the vessel breach impacts the walls directly. Because a detonation produces a shockwave (with a time scale on the order of milliseconds), any reflected pressure waves from surfaces complicates calculating the failure of the surrounding structures or connecting structures. However, if structural data are available, the effects of deflagration and non-reflected pressure pulses can be used for modeling a reprocessing facility accident [Rodriguez 95].

Determining the opening size in a wall or ceiling also requires knowledge of the structure geometry and material properties. Gases from an explosion travel through penetrations such as doors, windows, ventilation ducting or piping holes before or at the same time as the walls or ceiling fail.   In hazard calculations, there is a method called "explosion venting," which provides a pathway for discharging combustion gases during a deflagration to maintain pressures below the room damage threshold. This venting method is determined by the size of the vent in order to keep the equipment or other structures intact without damage. The fire standard NFPA 68, entitled "Standard on Explosion Protection by Deflagration Venting" describes this venting method. A discussion of explosion venting can be found in the fire handbook [NFPA 2008]. Similarly, a safe venting estimate for a red oil explosion (runaway reaction) has been discussed

elsewhere [Paddleford 1994]. In this reference, the venting is intended to prevent the red oil explosion because the accelerating temperature increase is a result of pressure build-up and run-away reaction rates at high-pressure conditions.

For a low-strength enclosure, the basic relation for a deflagration vent size is given as [NFPA 2008]:

$$A_v = C\, A_S / P_{red}^{1/2}$$

Equation 5.11

where A_v = required minimum vent area (m^2), C = venting equation constant, which is a function of the flame burning velocity (see Equation 5.12 below), A_S = enclosure surface area including ceiling, floor and walls (m^2), and P_{red} = maximum pressure developed in vented deflagration (bar$^{1/2}$). Note that this maximum pressure for this equation must be < 0.1 bar-g (gauge pressure). C for the low-strength enclosure is given as a function of burning velocity, v_b (cm/s):

$$C = 1.57 \times 10^{-5} v_b^2 + 1.57 \times 10^{-4} v_b - 0.0109$$

Equation 5.12

Equation 5.11 is restricted in application to cases in which the longest dimension of the enclosure (L_{max}), measured from the vent is [NFPA 2008]:

$$L_{max} \le c_L \frac{A_{CS}}{P}$$

Equation 5.13

where c_L = 12 for relatively unobstructed enclosures with quiescent gas mixtures or 8 for enclosures that have many internal obstructions or possibly highly turbulent gas mixtures, A_{CS} = the cross-sectional area normal to L_{max}, and P = perimeter of the cross section.

For the high-strength enclosures (P_{red} > 0.1 bar-g) [NFPA 2008], A_v is given by

$$A_v = \left[(0.127\, log_{10} K_G - 0.0567) P_{red}^{-0.582} + 0.175 P_{red}^{-0.572} (P_{stat} - 0.1)\right] V^{2/3}$$

Equation 5.14

where P_{stat} = vent deployment pressure (bar-g), and $K_G = 100\, S_u / 46$, which is a function of the combustible gas, and S_u = burning velocity (cm/s). Note that Equation 5.14 is for $L/D \le 2$, where L = longest dimension of an enclosure, and D is the equivalent diameter of the cross sectional area.

The challenge here is how to apply the above equations for the red oil explosions, and what burning velocity is appropriate for the red oil explosions.

Overpressure Model

Blast overpressure models discussed herein are based on shock wave propagation into the surroundings once the PVB has occurred [AIChE 2010]. Many of the analytical explosion methods, especially in the area of gas explosions in terms of detonation/deflagration (in relation to the flame speed) and confined explosions have been examined in [Louie 2004]. While direct simulation of structural damage is possible with CFD analyses, it is unclear whether a CFD code will be available or chosen for future applications given the limited availability of data to represent a facility. The analytical expressions presented below provide a means to estimate whether structural damage occurs. However, the methods are somewhat simplified and often may not be applicable for certain geometries.

In the last quarter century, a number of experiments and studies correlated PVB with solid explosive blast effects [Baker 1977, Coleman 1988 and Cain 1996]. Many experiments were performed in open air to observe PVBs due to pressurized gases or liquids [Cain 1996]. In the 1970s, the Baker model [Baker 1977] was developed for estimating the pressure wave effects in exploding propellant tanks and gas storage vessels. Experiments have confirmed Baker's model for determining overpressure and impulse as a function of distance from the tank rupture using both pressurized vessels and solid explosives [Baker 1977, Coleman 1988 and Cain 1996]. In the Baker-Strehlow model, Sach's scaling (or Energy scaling), is used to estimate the overpressure and impulse as a function of the scaled distance ($\bar{R}$) [AIChE 2010] as,

$$\bar{R} = R \left(\frac{P_0}{E_{exp}} \right)^{1/3} \qquad \text{Equation 5.15}$$

where P_0= ambient pressure, E_{exp}= explosion energy, and R= standoff distance. The scaled overpressure ($\bar{P}$), is defined as,

$$\bar{P} = \frac{P_s}{P_0} \qquad \text{Equation 5.16}$$

where P_s= side-on pressure.

A shock wave consists of positive and negative phases. Most of the time, only the positive phase is considered because it usually represents the largest change in pressure.

The scaled impulse ($\bar{i}$) is defined as:

$$\bar{i} = \frac{i \, a_0}{P_0^{2/3} E_{exp}^{1/3}} \qquad \text{Equation 5.17}$$

where i= impulse, and a_0= sound speed at the ambient condition.

To obtain the reflected overpressure (P_r) experienced by structures (e.g., walls), the following equations are needed [TIC-11268]:

$$\bar{P}_r = P_r / P_0 \qquad \text{Equation 5.18}$$

And

$$\bar{P}_r = 2 \bar{P} + \frac{(\gamma + 1) \cdot \bar{P}^2}{(\gamma - 1) \cdot \bar{P} + 2\gamma} \qquad \text{Equation 5.19}$$

As indicated in Equation 5.19, the scaled reflected overpressure ($\bar{P}_r$) is more than twice the scaled side-on pressure ($\bar{P}$). For air, γ= 1.4.

Based on the above scaled quantities, and solid explosive blast data, one could obtain P_s and i graphically. In this approach [Baker 1977], explosive blast curves for Pentolite for PVBs have been matched closely to gas burst data. However, this model is based on a spherical Pentolite

explosion in free air (which is away from the ground). To extrapolate these curves for cylindrical shaped explosives, correction factors must be applied [Ferradas 2006 and Geng 2011]. A recent paper has suggested that these correction factors should also take into account the length to diameter ratio of the cylindrical vessel, and the ratio of burst pressure to ambient pressure [Geng 2011]. For this study the use of the spherical burst overpressure and the inclusion of a reflecting surface (e.g., the ground) may be adequate to estimate the overpressure and impulse. In this approach, the burst pressure through the opening is assumed to be isotropic. Table 5-5 shows the correction factors to account for the situation near the ground. Graph lookup may not be an efficient method, so a recent paper has converted $\bar{P}$ and $\bar{i}$ estimates from this model into a set of equations that are functions of $\bar{R}$ in Table 5-6 for the spherical vessels.

Table 5-5   Scaled overpressure and impulse correction factors for vessels situated near ground [Ferradas 2006]

$\bar{R}$	Correction Factor	
	$\bar{P}$	$\bar{i}$
< 1	2	1.6
≥ 1	1.1	1

Table 5-6 Fitted $\overline{P_s}$ and $\bar{i}$ equations* for spherical vessels [Ferradas 2006]

$\bar{R}$ Interval	Scaled Overpressure Equation	Scaled Impulse Equation
$0.1 \le \bar{R} \le 0.2$	$\bar{P} = 1.25\,\bar{R}^{-1.92}$	$\bar{i} = 0.0164\,\bar{R}^{-1.97}$
$0.2 < \bar{R} \le 1.5$	$\bar{P} = 0.58\,\bar{R}^{-2.39}$	$\bar{i} = 0.0622\,\bar{R}^{-1.14}$
$1.5 < \bar{R} \le 1000$	$\bar{P} = 0.26\,\bar{R}^{-1.11}$	$\bar{i} = 0.0550\,\bar{R}^{-1.04}$

*Equations have included ground effects, see Table 5-6. The variables for these equations are defined in Equation 5.15, Equation 5.16 and Equation 5.17, and the units are in the MKS (Meter-Kilogram-Second) system. For example, the pressure is in Pascals, distance is in meters, and energy is in joules. Note $\bar{R}$ is measured from the center of the explosion, which means that this value cannot be zero because of the presence of the vessel.

For a typical steel vessel rupture [AIChE 2010], the rupture energy is on the order of 10^0 to 10^3 J. Energies as high as 10^8 to 10^{10} J were predicted for the TOMSK explosion accident (see Table 4-4). About 20% to 50% of the explosion energy is used in propelling fragments. These fragments include the pieces from the rupture vessel and liquid and solid materials from inside the vessel.

About 50% to 80% of the remaining energy may damage surrounding structures. Once the vessel fails, the expanded gas fills the accident room first and some energy is dissipated by this process. Any weak point in the accident room, such as unbolted equipment, ventilation ducting, windows, or doors can be propelled by the expanding gas. A portion of the remaining energy may fail walls or ceiling, or drive the gas to adjoining rooms. Eventually, the expanding gas

dissipates its remaining kinetic energy and attains equilibrium with the surrounding air. Therefore, both the shock overpressure and expanding gases from the PVB event should be accounted for when assessing damage to the facility.

One may assume that the pressure due to the explosion is on the order of the failure pressure of the vessel. This assumption is consistent with documented events. [Rodriguez 1995] discusses a forensic trace of the red oil explosion event at the TOMSK-7 radiochemical facility in 1993. In this event, the propelled ceiling plug of the room landed some distance from the site. From the calculations, the compartment with the vessel failure had a pressure of about 4-5 $\times 10^5$ Pa. Vessel failure pressure was determined to occur at about 30×10^5 Pa. This failure pressure corresponds to the failure property of the steel vessel. Note that the compartment pressure after PVB is based on the thermodynamic relation

$$P_1 V_1 = P_2 V_2$$
<div align="right">Equation 5.20</div>

where subscript 1 is the vessel and 2 is the compartment, and P and V are the pressure and gas volume, respectively.

Another approach for estimating the overpressure in an enclosure is to assume an ideal gas expansion at a constant volume with the final temperature as the adiabatic flame temperature from NUREG-1805 [Iqbal 04].

The overpressure may also be determined from high-explosive experimental data. In these experiments, the PVB is a result of a burst [Cain 1996]. Both compressed liquid and gas inside the vessel are used. Based on the burst, the explosion energy is calculated.

The explosion energy (E_{exp}) in Equation 5.15 needs to be determined to find the overpressure or specific impulse. A simple approach is to assume that this energy is equal to or greater than the design pressure if a material-defect or missile impact is the cause. These failure modes are not considered in this report.

The equations for the explosion energy are given below [Coleman 88]. Considering an ideal gas and isentropic condition, the explosion energy ($E_{exp,isen}$) is given by:

$$E_{exp,isen} = \frac{P_v V_v}{\gamma - 1} \cdot \left(1 - \frac{P_0}{P_v}\right)^{\frac{\gamma - 1}{\gamma}}$$
<div align="right">Equation 5.21</div>

where γ=heat capacity ratio of the gas, V_v= volume of the vessel, P_v= failure pressure of the vessel, and P_0= ambient pressure.

For adiabatic ideal gas conditions, the explosion energy ($E_{exp,adia}$) is given by:

$$E_{exp,adia} = \frac{P_v V_v}{\gamma - 1} \cdot \left(1 - \frac{P_0 \rho_v}{P_v \rho_0}\right)$$
<div align="right">Equation 5.22</div>

where ρ_v= gas density inside the vessel before failure, and ρ_0= ambient gas density. During PVB, gas from the vessel does not have time to transfer significant heat to the surrounding gas. This equation does not assume the expansion to be reversible, and it is not widely used because the value of ρ_v is needed.

For the isothermal ideal gas condition, the explosion energy ($E_{exp,iso}$) is given by:

$$E_{exp,iso} = P_v V_v \cdot ln\left(\frac{P_v}{P_0}\right)$$

Equation 5.23

Equation 5.23 assumes that the temperature does not change through the PVB. Also note that Equation 5.21 to Equation 5.23 can be applied only for the ideal gas situation. These equations need to be modified for non-ideal gas behavior.

Ideal gas behavior is adequate for most low pressure situations, less than about 1.03×10^7 Pa (1500 psi). To correct for real gas behavior, a compressibility factor, Z is defined as [Coleman 88]:

$$Z = \frac{V_{molar}}{V_{molar(ideal)}} = \frac{P}{R_{const}T} \cdot V_{molar}$$

Equation 5.24

where V_{molar} = molar volume (mole/unit volume) for the real gas, and R_{const} = gas constant. The term $\frac{R_{const}T}{P}$ is the molar volume for ideal gas, where T = temperature, and P = pressure. For ideal gases, Z = unity. For real gases, Z can be greater than or less than unity. Therefore, for real gases, Equation 5.21 to Equation 5.23 become, respectively:

$$E_{exp,isen} = \frac{P_v V_v}{Z(\gamma - 1)} \cdot \left(1 - \frac{P_0}{P_v}\right)^{\frac{\gamma-1}{\gamma}}$$

Equation 5.25

$$E_{exp,adia} = \frac{P_v V_v}{Z(\gamma - 1)} \cdot \left(1 - \frac{P_0 \rho_v}{P_v \rho_0}\right)$$

Equation 5.26

$$E_{exp,iso} = \frac{P_v V_v}{Z} \cdot ln\left(\frac{P_v}{P_0}\right)$$

Equation 5.27

Both the isothermal and isentropic expansion of a gas for predicting blast overpressure can be either real or ideal. However, both models tend to over predict the overpressure.

To utilize the extensive high explosive data for determining the blast overpressure [Cain 1996, Baker 1977, Geng 2011], this explosion energy is often converted to the equivalent mass of high explosive (such as TNT or Pentolite). TNT equivalents for various gases are given in Table 5-3. In addition to this table, an empirical correlation to convert the explosion energy from PVB or gas explosions (i.e., hydrogen or natural gas explosions in the room) to the TNT explosive equivalent mass, M_{TNT} is given by [Louie 04] as

$$M_{TNT} = \beta \frac{E_{exp}}{H_{TNT}}$$

Equation 5.28

where β = a TNT equivalence factor, yield factor, or efficiency factor, and H_{TNT} = TNT blast energy. A value of H_{TNT} is 4.68 MJ/kg [AIChE 10]. Note that β in Equation 5.28 is for combustion gases and varies from 0.03 to 1.0 [Louie 04].

Unlike the vapor cloud explosion condition, the assumption of a PVB is that the explosion occurs inside a vessel, so that β is unity. Once M_{TNT} is estimated, then it can be used as input to the code BLASTX.

5.2.2   Release Due to Nuclear Criticality

A release can occur due to an inadvertent criticality event (often referred to as a nuclear excursion). For a closed vessel to be ruptured, the fission energy of the criticality event must be sufficiently large to overcome the design pressure of the vessel.   If this energy is sufficient to rupture the vessel, the remaining energy may expel solution from the breached vessel, and some of this expelled solution can form aerosols and vapors. This accident has a similar aerosol formation process as discussed previously for explosions. However, fission energy is usually distributed uniformly over the volume, unlike a chemical explosion, which may be concentrated within a smaller region of a vessel. Moreover, criticality events do not create gaseous products from a chemical reaction, but only from change of phase due to an increase in temperature. In view of these arguments, a criticality event can be modeled similarly as a chemical explosion in terms of the formation of vapors and aerosols, with the exception that gas-phase products are not directly formed from the fission energy.

For an inadvertent criticality condition, the explosion energy should be calculated based on the total number of fissions. If the fission energy is large enough to exceed the failure pressure of the vessel, then Equation 5.21 to Equation 5.27 are not needed.

5.2.3   Fires

A limited number of experiments have been performed for liquid combustion in the context of reprocessing solvents, which involve both nitric acid and organic solutions [NUREG/CR-4736]. In one set of experiments, small volumes of 30% tributyl phosphate (TBP) in normal paraffin hydrocarbon (NPH, a kerosene-like diluent used in fuel reprocessing) were burned.   Ignition was provided by a methane flame located above a beaker containing the liquids. The NPH was the only combustible material in the experiment. The acids were included to see if fire and heating would cause migration of the fission products or transuranic elements across the phase boundaries.

An empirical correlation was developed using the combustible liquid of 30% TBP/kerosene for the rate of release of airborne uranium release rate with an aerodynamic mass mean diameter (AMMD) of 0.6 µm, and geometric standard deviation (GSD) of 3.1 [NUREG-1320]. This correlation is given as

$$\dot{m}_{rel} = 1.38\, S_r w_{rad}$$

Equation 5.29

where $\dot{m}_{rel}$= mass release rate of radioactive particles (g/s), w_{rad} = mass of the radioactive material in the solution (g), and S_r= smoke release dimensionless rate (s^{-1}). S_r is given as [NUREG-1320]:

$$S_r = \frac{\gamma_s \left(\dot{q}''_{rr} + \dot{q}''_{fc} + \dot{q}''_{fr} \right) A_b}{h_c} m_f$$

Equation 5.30

where γ_s= fractional yield of smoke (for TBP/kerosene: 0.002 to 0.087), $\dot{q}''_{rr}$= surface radiation heat loss (for kerosene: 8 kW/m^2), $\dot{q}''_{fc}$= flame convective heat flux (for kerosene: 11 kW/m^2),

$\dot{q}''_{fr}$ = flame radiative heat flux (for kerosene: 14 kW/m²), A_b = burning surface area of the combustible liquid (m²), h_c = heat required to generate a unit mass of fuel vapors (for kerosene: 1.5 kJ/g), and m_f = mass of the combustible liquid.

In addition to this empirical correlation associated with combustible liquids, airborne release and aerosol size are provided in Table 5-7 and Table 5-8 for combustible liquids.

Table 5-7 Summary of the airborne release in combustible liquid [NUREG/CR-4736]

Experiment	Airborne Release Fraction	Notes**
Pure acid/organic with uranium	0.004-0.0056	100 mL organic liquid was placed on top of 100 mL of acid in a metal beaker. External heat was supplied. Substitution of a glass beaker would only delay the acid boil over. Approximately 40-60% of organic layer remained unburned. The amount of smoke generated is 0.0076 to 0.0189, and the burn time ranges from 27.5 to 53.3 minutes.
Acid with fission products/organic with uranium (30% TBP in NPH*)	0.025-0.027 uranium	The presence of fission products in the acid phase seems to enhance uranium release from the organic phase. The amount of smoke generated is from 0.0288 to 0.0318 and the burn time ranges from 24.8 to 34 minutes.
Acid with fission products and uranium/pure organic (TBP in NPH)	0.06-0.071 uranium	These experiments did burn to solid residue, with the initial solution volume of 100 mL in a metal beaker.
Acid with fission products and uranium/pure organic (TBP in NPH)	0.002 uranium	This experiment did not burn to solid residue. Its initial solution volume was 150 mL of acid with 50 mL of organic in a glass beaker with a heat pump to provide external heat flux. The smoke release is from 0.0644 to 0.0891, and burn time ranges from 40 to 65 minutes.
Acid with fission products and uranium/organic with uranium	0.008-0.016 uranium	Experiments involved solutes in both the organic and acid layers. Experiments did not burn to a solid residue. Note that one experiment has provided aerosol size: AMMD at 0.6 µm, GSD of 3.1, and 99% < 10 µm. The smoke release is from 0.0303 to 0.0651 and burn time ranges from 51 to 57.3 minutes.

*NPH has the following properties: specific gravity at 15.6 ˚C is 0.76, viscosity at 25 ˚C is 1.8 centipoise, and flash point at 70 ˚C.
** The lower burn time value should be used to estimate the release rate.

Table 5-8 Source term for burning of combustible liquid [NUREG-1320]

Release Mechanism	Airborne Release Fraction	Airborne Particle Size	
		AMMD (µm)	GSD
Burning of contaminated combustible liquids			
U or Pu liquid (Combustible liquid is spilled over large amount of radioactive material, then	0.114	2.4	3.8
ignited)	0.003	2.4	3.8
U or Pu nitrate	0.0077	2.4	3.8
Non-volatiles other than U or Pu	0.01	2.4	3.8
Semi-volatiles	0.843	2.4	3.8
Volatiles			
Heating of unpressurized radioactive liquids			
Pre-boiling	1.06×10^{-10}/s	20.0	1.5
Boiling	4.76×10^{-7}/s	20.0	1.5

Fires also induce a buoyant plume which affects the transport of airborne particles. Fire codes are available and the ones listed below can estimate the transport of energy and mass, and characterize plume rise. However, aerosol generation and dynamics are not determined with these codes. The following codes could be used to perform estimates of burn rate, energy generation, fire duration, and plume rise as inputs to another code. The fire codes include: CFAST (Consolidated model of Fire and Smoke Transport) and FDS (Fire Dynamics Simulator). The descriptions of these codes are briefly provided below:

> CFAST simulates impact of fires and smoke in a specific building environment. A two-zone fire model is used to calculate evolving distribution of smoke, fire gases, and temperature throughout rooms of a building during a fire.

> FDS models the fire-driven fluid flow. This code solves numerically a form of the Navier-Stokes equations appropriately for low-speed, thermally-driven flow, with an emphasis on smoke and heat transport from fires.

5.2.4   Spills

Spill accidents in a fuel reprocessing facility are mainly due to leaks from valves and lines, and cracks in tanks. The leaks are due to corrosion, overpressure, and mechanical impact. A spill due to overpressure is called a "pressurized spill" and is discussed later in this section.

In terms of the radionuclide behavior, particularly in creating an airborne release, a free-fall spill usually exhibits break-up of the liquid into drops due to the instability and shear stress at the surface of the liquid. Once the liquid/drops hit a hard, unyielding surface to create splashing, drops are broken into smaller ones and some of these drops are resuspended. The amount of aerosol created by spills is less than that from more energetic events such as fire, criticality, and explosion accidents. For a non-pressurized spill, the energy source that creates suspended drops is gravitational potential.

Non-Pressurized Spill

NUREG/CR-4997 documents the performance of a number of free-fall spill experiments involving powders and solutions. The properties of the experimental solutions compared with

water are shown in Table 5-9. Because it was impractical to sample the entire air volume immediately after the spill, a correction was made to account for evaporation effects in the data. For example, uranine solution particles were most affected by evaporation because uranine solution contains only 1% non-volatile solids; whereas, the UNH solutions contained 28% non-volatile solids and a large percent of lower volatility HNO_3.

Table 5-9 Comparison of properties of water and experimental liquids*

Material	Density (g/cc)	Viscosity (centipoise)	Surface Tension (dyne/cm)	Vapor Pressure (mm Hg)
Water	0.997	0.89	72.0	23.8
UNH [a]	1.54	1.70	66.4	13.6[b]
Uranine [c]	0.993	0.79	52.1	23.7
Sucrose [d]	1.0-1.3	1.3-47.1	67.0-83.0	
Slurry	1.1-1.4	1.3-3.1	58.2-68.4	

*at 25 ˚C, and adapted from Table 3.2 of [NUREG/CR-4997].
[a] Uranyl nitrate hexahydrate solution in nitric acid with 208.7 g uranium/liter (0.877 Molar), 8.4 Molar HNO_3.
[b] Calculated
[c] 10 g/liter uranine in water
[d] Varied from 0 to 60% sucrose

Table 5-10 shows the airborne release data for the free-fall spill experiments. The initial data are generally higher than the impactor data, since the initial data account for evaporation and settling (see [NUREG/CR-4997] for more details about the data corrections).

Table 5-10   Experimental results on liquid spills with correction [NUREG/CR-4997]*

Applied Condition	Initial [a]	Measured [b]	Initial/Measured
All Spills[1]			
Average AMMD, μm	21.5	8.9	2.4
Average GSD[c]	7.3	5.4	1.4
Average Airborne Fraction	28.5×10^{-6}	25.5×10^{-6}	1.1
UNH Spills[2]			
Average AMMD, μm	27.2	20.1	1.4
Average GSD[c]	5.9	4.5	1.3
Average Airborne Fraction	9.3×10^{-6}	7.9×10^{-6}	1.2
Uranine Spills[3]			
Average AMMD, μm	27.0	7.3	3.7
Average GSD[c]	3.0	2.9	1.0
Average Airborne Fraction	56.2×10^{-6}	50.7×10^{-6}	1.1
Sucrose Spills[4]			
Average AMMD, μm	11.0	2.7	4.1
Average GSD[c]	14.2	9.4	1.5
Average Airborne Fraction	10.6×10^{-6}	9.7×10^{-6}	1.1
Slurry Spills[4]			
Average AMMD, μm	15.8	3.1	5.1
Average GSD[c]	10.1	6.7	1.5
Average Airborne Fraction	23.3×10^{-6}	20.7×10^{-6}	1.1
Low Surface Tension Spills[5]			
Average AMMD, μm	20.2	6.5	3.1
Average GSD[c]	3.7	4.2	0.9
Average Airborne Fraction	45.9×10^{-6}	41.5×10^{-6}	1.1

*The experiments included a spill height from 1 to 3 m onto a dry stainless steel floor, liquid volume of 125 to 1000 ml. Liquids spilled included slurries and solutions of varying viscosities. The spill experiment is for a single spill with a spill diameter (of spill container) from 6.2 to 12.4 cm.
[a] Recalculated to account for any evaporation and settling effect.
[b] Experimental impactor data
[c] Geometric standard deviation, lognormal distribution
[1] If the liquid cannot be specified, use the data in this set
[2] It represents heavy metal, aqueous solutions with densities > 1.2 g/cc
[3] It contains less than 1% non-volatiles
[4] See Table 5-9 for properties identified
[5] Surface tension is 33.3 dyne/cm, viscosity is 1.14-1.24 centipoise, and density of 1.004 g/cc.

NUREG/CR-4997 includes empirical correlations for the airborne release fraction (ARF) for liquid spill experiments. Equation 5.31 includes an evaporation and settling correction:

$$ARF = 8.9 \times 10^{10} Arch^{0.55}$$ Equation 5.31

where $Arch$ is the Archimedes number, which is defined as

$$Arch = \frac{\rho_{air}{}^2 H^3 g}{\mu}$$ Equation 5.32

where ρ_{air}= air density (g/cc), H= spill height (cm, extrapolating above 3 m may not be reasonable), g = gravity (981 cm/s^2) and μ= solution viscosity (poise). For a very low-density aqueous solution, the right hand side of Equation 5.31 should be multiplied by 3.

An empirical correlation similar to Equation 5.31, but which includes the Froude number and dimensionless density, can be used to estimate ARF

$$ARF = 6.31 \times 10^{-6} Fr^{0.35} Arch^{0.45} \left(\frac{\rho_{air}}{\rho_s}\right)^{2.2}$$ Equation 5.33

where Fr = Froude number and ρ_s = density of the solution. Fr in Equation 5.33 is given by

$$Fr = \frac{v^2}{gR}$$ Equation 5.34

where v = impact velocity ($v = \sqrt{2 g H}$, cm/s) and R = radius of equivalent sphere of liquid spilled (cm).

Note that Equation 5.34 includes a dependence on impact velocity and drop size, which Equation 5.31 does not. Also, note that the data in Table 5-10 and the above correlations are only applicable for a single spill event. For spills where the drop diameter is < 8 mm, and the spill height > 4.26 m, the results of the correlations are likely to be conservatively high because of the way velocity is computed. In the case of continuous spills, where a falling stream encounters less air drag than individual droplets, the correlations may be non-conservative.

As described in [NUREG/CR-4997], attempts have been made to correlate the lognormal aerosol size parameters, *AMMD* and *GSD*. Only the slurry spill data yields a correlation with a least square greater than 60%, and it is given as [NUREG-1320]:

$$AMMD \ (\mu m) = 2.3 \times 10^{-8} \frac{We^{1.9}}{Re^{0.16}}$$ Equation 5.35

Where We= Weber number and Re = Reynolds number and these are given as:

$$We = \frac{\rho_s d v^2}{\epsilon_t}$$ Equation 5.36

where d= diameter (cm), ϵ_t = liquid surface tension (dyne/cm), and v = velocity (cm/s), which is $\sqrt{2 g H}$, and

$$Re = \frac{\rho_s v d}{\mu}$$ Equation 5.37

A sample calculation in NUREG/CR-4997 evaluates a hypothetical leak in a solvent line in a reprocessing plant. The solvent is 30% tributyl phosphate (TBP with kerosene as a diluent). This solvent has a viscosity of 0.0207 poise. It contains 0.1 Molar UNH and 1 Molar HNO_3. It is assumed that the leak is at 25 ˚C and falls 366 cm (12 ft) to the floor.

From Equation 5.32 with air density of 0.00155 g/cc,

$$Arch = \frac{(0.00155 \ g/cc)^2 (366 \ cm)^3 (981 \ cm/s^2)}{\left(0.0207 \frac{g}{cm \cdot s}\right)^2} = 2.7 \times 10^8$$ Equation 5.37

Using Equation 5.32, the ARF is 3.86 x 10^{-5} and from Table 5-10, $AMMD$ = 21.5 µm and GSD = 7.3.

Pressurized Spill

A pressurized spill is a release caused by a pressure that provides an initial velocity to the ejected liquid. It is expected that the ARF is higher than for the free-fall spill case.

A series of experiments were performed on pressurized releases of solutions [NUREG/CR-3093, NUREG/CR-4779]. These experiments were conducted using so-called Pressurized-Airborne Release Equipment (PARE) inside a cylindrical enclosure called the Radioactive Aerosol Release Tank. As shown in Figure 5-8, the chamber is about the size of a small room. Also indicated in this figure, the release is upward toward the ceiling of the chamber. This configuration should provide an upper bound on the ARF. The size of the opening of the rupture disk limits the amount of the liquid expelled. Aerosol is collected by impactors and filters that are located near the curved walls of the cylinder. Since PARE is designed for pressurized releases of powders, PARE was modified to measure liquid releases as shown in Figure 5-9. The initial set of the experiments were conducted using air as the pressurized gas at pressures ranging from 50 to 500 psig. The initial solution of uranine and uranyl nitrate had volumes between 100 cm^3 and 350 cm^3 [NUREG/CR-3093]. Because of concern that the ARF may be a function of the amount of gas dissolved in the liquid, additional tests were performed with CO_2 [NUREG/CR-4779]. As shown in Table 5-11, the solubility of CO_2 in uranine solution is 50 times that of air. Table 5-12 provides the airborne release data for the comparison of air and CO_2 for pressurized uranine solution releases. As shown in this table, using air as the pressurized gas tended to yield a lower airborne ARF value in comparison with CO_2.
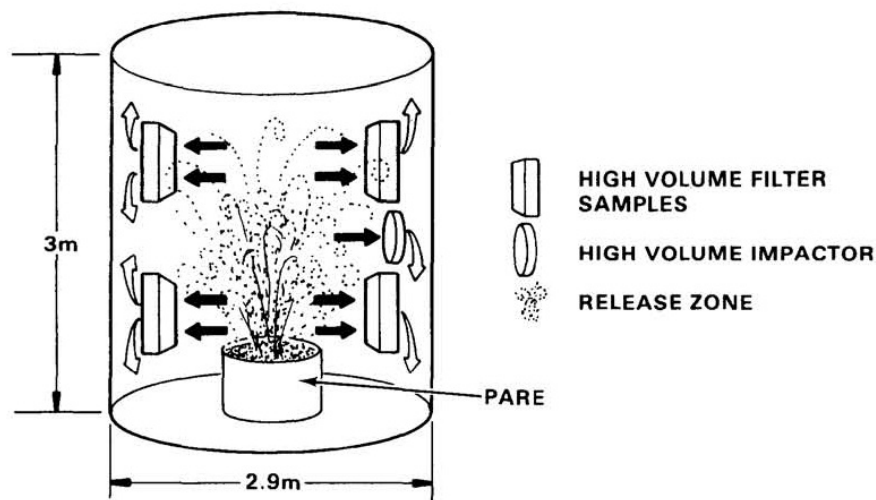
Figure 5-8   Pressurized radioactive aerosol tank configuration for measuring the pressurized release of solution from PARE [NUREG/CR-3093]
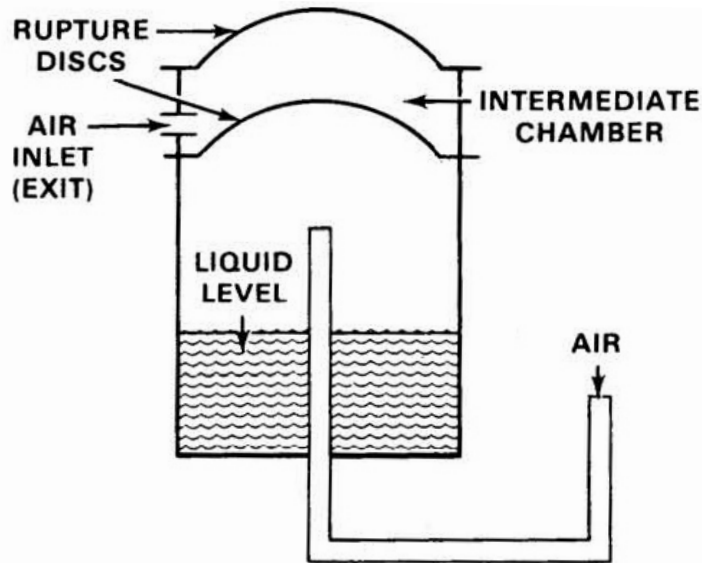
Figure 5-9   PARE modification for pressurized liquid release experiment [NUREG/CR-3093]

Table 5-11   Estimates of mole fraction of the pressurized gases dissolved in uranine solution* [NUREG/CR-4779]

Pressure (psig)	Pressurized Gas	Mole Fraction Dissolved
500	Air CO_2	4.2×10^{-4}** 2.0×10^{-2}
250	Air CO_2	2.3×10^{-4} 1.3×10^{-2}
50	Air CO_2	4.8×10^{-5} 2.5×10^{-3}

*Assume solubility of gases the same as in water. This solution contains a uranium concentration of 10 g/liter.
** Air value calculated assuming 78% N_2, 21% O_2; 1% inert (not included)

Table 5-12   Airborne release for pressurized release of uranine solutions [NUREG/CR-4779]

Chamber Pressure (psig)	350 cc Volume						100 cc Volume					
	Air			CO_2			Air			CO_2		
	ARF	AMMD (μm)	GSD	ARF	AMMD (μm)	GSD	ARF	AMMD (μm)	GSD	ARF	AMMD (μm)	GSD
500	0.05	4	2.7	0.13	3.3	11	0.15	4	3.5	0.22	2.7	5.5
250	0.01	4	2.3	0.039	2.3	5.2	0.06	3	3.8	0.50	2.1	2.6
50	0.0008	2	3.5	0.014	1.5	3.8	0.005	2	4	0.006	1.9	3.3

In addition to a pressurized release, a flashing-spray release experiment was also conducted for an uranine solution at the same concentration. In this experiment, the rupture pressure was the vapor pressure of superheated liquid [NUREG/CR-4779]. Figure 5-10 shows the measured and predicted temperatures of the flashing-spray release experiment. The ARF data for this flashing-spray experiment are shown in Table 5-13.
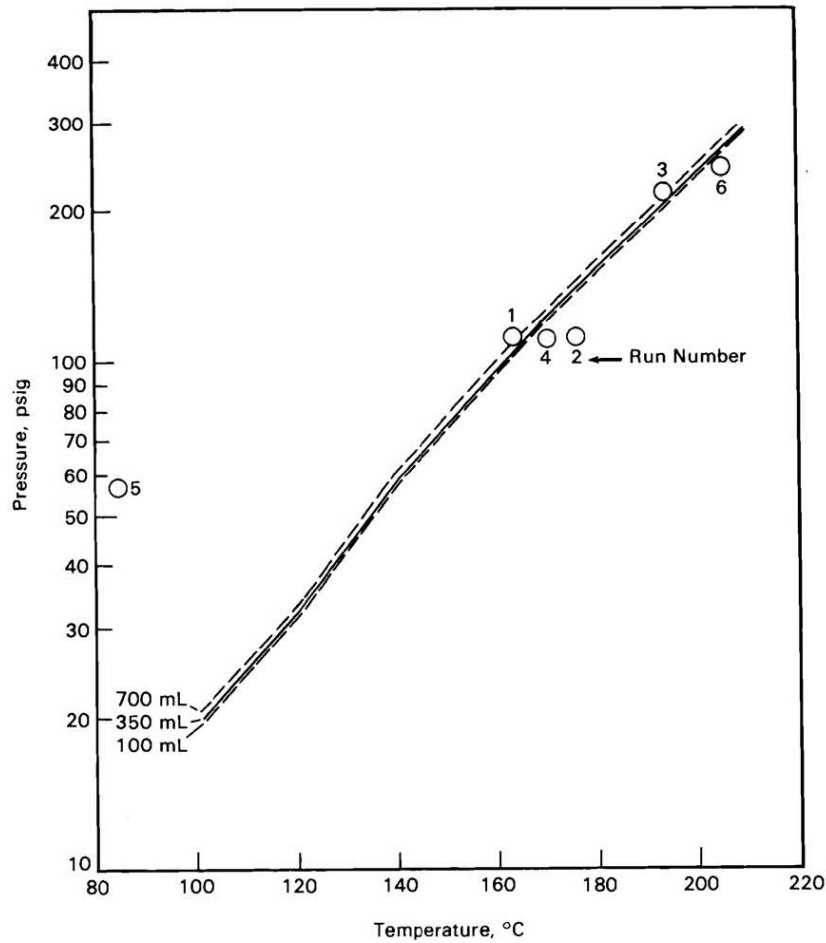
Figure 5-10   Measured and predicted temperatures for flashing-spray releases [NUREG/CR-4779]

Table 5-13   Airborne release for flashing-spray releases of uranine solutions [NUREG/CR-4779]

Chamber Pressure (psig)	700 cc Volume			350 cc Volume			100 cc Volume		
	ARF	*AMMD (µm)*	*GSD*	*ARF*	*AMMD (µm)*	*GSD*	*ARF*	*AMMD (µm)*	*GSD*
240				0.0496	6.25	2.7			
124	0.0151	5.9	2.6	0.0222	7.7	2.7	0.0851	6.4	3.3
57				0.0089	8.4	5.6			

Unlike the uranine solution, UNH solution contains more non-volatiles in solution, and the density is about 1.54 g/cc. The ARF results for pressurized release of UNH solution using air are given in Table 5-14.

Table 5-14   Airborne release for the pressurized UNH solutions in air [NUREG/CR-3093]

Chamber Pressure (psig)	350 cc Volume			100 cc Volume		
	ARF	*AMMD (μm)*	*GSD*	*ARF*	*AMMD (μm)*	*GSD*
500	0.009	16	3.4	0.01	14	2.7
250	0.005	29	4.4	0.0009	13	3.6
50	0.0002	4	5.0	0.0003	7	6.1

The empirical correlation for ARF during a pressurized release reported in NUREG-1320 is

$$ARF = 0.3 \, x_g^{0.91}$$

Equation 5.38

where x_g = mole fraction of pressurizing gas or vapor. The *AMMD* associated with Equation 5.31 is 6.8 μm and the *GSD* is 3.2.

Similar to the spill cases in the previous section, a correction was made to the measured data set for evaporation and settling [NUREG-1320]. With this correction, Equation 5.39 becomes

$$ARF = 0.33 \, x_g^{0.91}$$

Equation 5.39

A comparison of the aerosol size distribution data for liquid pressurized releases with and without the evaporation and settling correction is given in Table 5-15. As concluded in [NUREG/CR-3093], measured ARF values were a function of pressure, but were relatively independent of the volume of the experimental configuration in Figure 5-8.

Table 5-15   Comparison of the measure and correction data of aerosol size distribution for liquid pressurized release [NUREG-1320]

Experiment	Measured Data		Evaporation/Settling Corrected Data	
	AMMD (μm)	GSD	AMMD (μm)	GSD
All Data	7.3	3.9	12.0	6.6
Uranine/Air	3.3	3.2	12.4	4.5
UNH/Air	13.6	4.2	8.4	11.1
Uranine/CO_2	2.3	5.2	9.3	5.6
Flashing Sprays	6.8	3.2	21.0	3.0

5.3   Basic Principles Approach to Determining Releases

As discussed in the previous section, progress has been reported for determining the factors in the five-factor formula used to determine the source term. For convenience, the formula is

MAR $\times$ DR $\times$ ARF $\times$ RF $\times$ LPF,

which in expanded form is

Material At Risk $\times$ Damage Ratio $\times$ Airborne Release Fraction $\times$ Respirable Fraction $\times$ Leak Path Factor.

For this application, the MAR is the total mass of solution inside a unit being considered. Deposition and filtration effects may significantly reduce the LPF below unity, and can be determined with transport codes. However, estimates of DR, ARF, and RF are very difficult to obtain. In particular, scaling from 350 cm^3 beaker-size experiments to cubic meter reprocessing units is questionable. Furthermore, the particle size distribution is not determined by the five-factor formula approach, neither is the variation in radiological content with particle size. The particle size distribution and radiological content greatly influences transport within facilities, dispersion in the atmosphere, and health consequences. For the five-factor formula approach, the respirable fraction (RF) is used as a rough measure to encompass these effects. Furthermore, there are no experiments that we found on the scale used in reprocessing facilities. Instead, suggestions for ARF and RF given in the previous section are for isolated effects, and these are generally not on the scale of interest.

As part of this program, a basic principles approach is considered that not only provides the DR, ARF, and RF, but also provides the particle size distribution and radiological content of the aerosol with particle size, while simultaneously determining the failure of the containing vessel. The approach is to model from basic principles the coupled problems of explosion dynamics, liquid solution release and break up, and structural vessel rupture and failure.   These phenomena are simulated by coupled structural and fluid mechanics codes, including a drop break-up model that is stable and accurate. All these codes have been developed under a common architecture called Sierra to facilitate the multi-physics coupling [Edwards 2002].

Sandia's Presto structural mechanics code, which is a Lagrangian, three-dimensional, explicit, transient code is used to solve the structural problem with large deformations and short time scales. Radionuclide liquid particle dispersion is modeled with Smoothed Particle Hydrodynamic (SPH) elements, which are coupled to the explosive and the processing vessel walls [Monaghan 2005]. SPH allows for radionuclide solutions to both impart momentum to solid structures and for the solutions to be dispersed upon ejection from the vessel. The solid walls and equipment in the vessel are modeled with structural finite elements because these elements may deform and separate, but they do not get atomized by the explosive. After the fluid has been ejected and drops on the order of centimeters to millimeters have separated, the system is then modeled with Sandia's Fuego fluid mechanics code. Fuego is a low Mach number control-volume finite element code that solves the Navier-Stokes equations for the flow induced by the explosion and the ejected drops. Drop break-up is modeled using the Taylor Analogy Break-up (TAB) model [O'Rourke 1987; Desjardin 2002] employing a Lagrangian transport framework coupled to the Eulerian gas phase solvers. The Presto/Fuego coupling is one-directional in that the structural code with SPH is run first, and then the fluids code with the TAB model is used to determine the aerosol particle size distribution and concentration. Typically, the fluid is initially modeled with several hundred thousand SPH particles, and then the final aerosol has orders of magnitude more particles that result from the break-up of SPH particles. These are demanding calculations that require a parallel machine running for several days, but for the first time reprocessing accidents can be simulated in detail from first principles instead of relying on limited and conservative correlations for the explosion, structural deformation of the reprocessing unit, and liquid aerosolization that are often limited to only one or a few of the phenomena.

6.   REFERENCES

[AGNS 1975]   **Allied-General Nuclear Services**, "Barnwell Nuclear Fuel Plant – Separations Facility Final Safety Analysis Report," Amendment 3, February 1975.

[AIChE 2010]   **American Institute of Chemical Engineering**, "Guidelines for Vapor Cloud Explosion, Pressure Vessel Burst, BLEVE and Flash Fire Hazards," 2nd Edition, Wiley, 2010.

[ANS-8.1]   **ANSI/ANS-8.1-1998**, "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," American Nuclear Society, 1998.

[Bader 2011]   S. Bader et al., "The COEX Process: Recycling of UNF in the U.S. as Proposed by AREVA," NRC-DOE Seminar, May 26, 2011.

[Baehr 1989]   W. Baehr, "Industrial Vitrification Processes for High-Level Liquid Waste Solutions," IAEA Bulletin, 4/1989, page 43-46.

[Baker 1977]   W.E. Baker et al., "Workbook for Predicting Pressure Wave and Fragment Effects of Exploding Propellant Tanks and Gas Storage Vessels," NASA Contractor Report 134906, September 1977.

[Ballinger 1987]   **NUREG/CR-4779 (PNL-6065)**, M.Y. Ballinger et al., "New Data for Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, May 1987.

[Ballinger 1988]   **NUREG/CR-4997 (TI99 004796)**, M.Y. Ballinger et al., "Method for Describing Airborne Fractions of Free Fall Spills of Powders and Liquids," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, January 1988.

[Bastin 2008]   C. Bastin, "We Need to Reprocess Spent Nuclear Fuel, And Can Do It Safely, At Reasonable Cost," http://21stcenturysciencetech.com/Articles%202008/Summer_2008/Reprocessing.pdf

[Beall 1997]   **NISTIR 6030**, K.A. Beall, editor, "Thirteenth Meeting of the UJNR Panel on Fire Research and Safety," U.S. Department of Commerce, June 1997.

[Bloom 1993]   **Y/ENG/SE-118**, S.G. Bloom and W.H. Moon, Jr., "Analysis of Explosion-Induced Releases of Toxic Materials at an Environmental Restoration Project," Paper submitted to Winter Annual Meeting ASME, New Orleans, La, November 28-December 3, 1993.

[BNL 2011]   **NUREG/CR-7168**, G. Martinez-Guridi et al., "Regulatory Approaches for Addressing Reprocessing Facility Risks: an Assessment," prepared by Brookhaven National Laboratory, U.S. Nuclear Regulatory Commission Washington, DC, February 2015.

[BNWL-931]   J. Mishima et al., "Plutonium Release Studies – IV. Fractional Release from Heating Plutonium Nitrate Solutions in a Flowing Air Stream," BNWL-931, Pacific Northwest Laboratory, November 1968.

[Brown 2009a]   A. L. Brown, "Impact and fire modeling considerations employing SPH coupling to a dilute spray fire code," Proceedings of the 2009 ASME Summer Heat Transfer Conference, HT2009-88493, ASME SHTC-2009, San Francisco, California, USA, July 19-23, 2009.

[Brown 2009b]   **IMECE2009-11675**, A. L. Brown and R. A. Jepsen, "An improved drop impact model for Lagrangian/Eulerian coupled codes," Proceedings of the 2009 ASME IMECE Conference ASME IMECE-2009, Lake Buena Vista, Florida, USA, November 13-19, 2009.

[Brown 2010]   **IHTC14-23067**, A. L. Brown and G. J. Wagner, "Fluid spread model validation for emerging liquid tank impact predictive methods," Proceedings of the 2010 ASME IHTC Conference, ASME IHTC14-2010, Washington, DC, USA, August 8-13, 2010.

[Brown 2011]   A. L. Brown, G.J. Wagner, and K.E. Metzinger, "Impact, Fire and Fluid Spread Code Coupling for Complex Transportation Accident Environment Simulation," *Journal of Thermal Science and Engineering Applications*, Accepted for publication, October 2011.

[Cadwallader 2005]   **Idaho National Engineering and Environmental Laboratory**, L. Cadwallader et al., "Summary of Off-Normal Events in US Fuel Cycle Facilities for AFCI Applications," INEEL/EXT-04-02557, September 2005.

[Cain 1996]   **45SPW-TR-96-01**, M.R. Cain, "Pressure Vessel Burst Test Study," General Physics Corporation, November 1996.

[Campbell 1990]   D.O. Campbell and W.D. Burch, "The Chemistry of Fuel Reprocessing: Present Practices, Future Trends," *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 142 No.1, page 303-320, 1990.

[Chan 1989]   **NUREG/CR-3037 (PNL-4532)**, M.K.Chan et al., "User's Manual for FIRN – A Computer Code to Estimate accidental Fire and Radioactive Airborne Releases in Nuclear Fuel Cycle Facilities," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, February 1989.

[Coleman 1988]   **ESMC-TR-88-03**, M. Coleman et al., "A Review of Energy Release Processes from the Failure of Pneumatic Pressure Vessels," General Physics Corporation, August 1988.

[Cooperstein 1974]   R. Cooperstein et.al, "Preliminary Environmental Analysis of a Generic Fuel Reprocessing Facility," EPA-520/3-75-003, SAI Services, May 1974.

[Croff 2008]　　　　**NUREG-1909**, A.G. Croff et al., "Background, Status, and Issues Related to the Regulation of Advanced Spent Nuclear Fuel Recycle Facilities," ACNW&M White Paper, U.S. Nuclear Regulatory Commission, Washington, DC, June 2008.

[Davydov 2003]　　　M.N. Davydov and V.K. Kedrinskii, "Two-Phase Models of Formation of Cavitating Spalls in a Liquid," Translated from Prikladnaya Mekhanika I Tekhnicheskaya Fizika, Vol. 44, No. 5, pp. 71-98, September-October, 2003.

[Desjardin 2002]　　**SAND2002-3419**, P.E. Desjardin and L.A. Gritzo, "A Dilute Spray Model for Fire Simulations: Formulation, Usage and Benchmark Problems," Sandia National Laboratories, Albuquerque, NM, October (2002).

[Dey 2005]　　　　　P.K. Dey and N.K. Bansal, "Spent fuel reprocessing: A vital link in Indian nuclear power program," *Nuclear Engineering and Design*, 236, page 723-729, 2006.

[DNFSB-T33]　　　　**DNFSD/TECH-33**, "Control of Red Oil Explosions in Defense Nuclear Facilities 2003," Defense Nuclear Facilities Safety Board Technical Report, November 2003.

[DOE 1992]　　　　　**DOE/TIC-11268**, "A Manual for the Prediction of Blast and Fragment Loadings on Structures," U.S. Department of Energy, July 1992.

[DOE 1994]　　　　　**DOE HDBK-3010-94**, "Airborne release fractions/rates and respirable fractions for nonreactor nuclear facilities," DOE Handbook, U.S. Department of Energy, 1994.

[DOE 1997]　　　　　**DOE/RL-97-63**, "Accident investigation board report on the May 14, 1997 Chemical Explosion at the Plutonium Reclamation Facility," Hanford Site, Richland, Washington, July 26, 1997.

[DOE 1999]　　　　　**U.S. Department of Energy**, Waste Vitrification Systems Lessons Learned, March 1999. Richland, Washington, July 26, 1997.

[DOE 2003]　　　　　**DOE HDBK-1169-2003**, "Nuclear Air Cleaning Handbook," DOE Handbook, U.S. Department of Energy, 2003.

[DOE 2004]　　　　　**U.S. Department of Energy,** "MELCOR Computer Code Application Guidance for Leak Path Factor in Documented Safety Analysis – Final Report," May 2004.

[DOE 2005]　　　　　**DOE/EH-0697**, "Significant Radioactive Leak at Sellafield due to Operational Complacency," Environment, Safety and Health Bulletin, Issue No. 2005-11, August 2005.

[DOE/EH 1998]　　　**DOE/EH-0555**, D.G. Harlow et al., "Technical Report on Hydroxylamine Nitrate," U.S. Department of Energy, February 1998.

[Edwards 2002]   **SAND2002-3616**, H. C. Edwards, "SIERRA Framework Version 3: Core Services Theory and Design," Sandia National Laboratories, November 2002.

[Elliott 1994]   **PNL-9822**, M.L. Elliott et al., "Prelimary Melter Performance Assessment Report," Pacific Northwest laboratory, August 1994.

[Ferradas 2006]   E.G. Ferradas et al., "Characteristic Overpressure-Impulse-Distance Curves for Vessel Burst," Progress Safety Progress, Volume 25 No.3, American Institute of Chemical Engineering, Page 250-254, September 2006.

[Geng 2011]   J. Geng et al., "Pressure Vessel Burst Directional Effects," Proceedings of the ASME 2011 Pressure Vessels & Piping Division Conference, July 17-21, 2011.

[Gephart 2003]   **PNNL-13605**, R.E. Gephart, "A Short History of Hanford Waste Generation, Storage, and Release," Rev.4, Pacific Northwest National Laboratory, October 2003.

[GFMR 2010]   "Global Fissile Material Report 2010: Balancing the Books: Production and Stocks," Fifth Annual Report of the International Panel on Fissile Materials, International Panel on Fissile Materials, 2010.

[Halverson 1986]   **NUREG/CR-4593 (PNL-5839)**, M.A. Halverson and J. Mishima, "Initial Concepts on Energetics and Mass Releases During Nonnuclear Explosive Events in Fuel Cycle Facilities," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, September 1986.

[Halverson 1987]   **NUREG/Cr-4736 (PNL-5999)**, M.A. Halverson, et.al, "Combustion Aerosols Formed During Burning of Radioactively Contaminated Materials – Experimental Results," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, March 1987.

[Ho 1988]   V. Ho et al., "COMPBRN III – A Fire Hazard Model for Risk Analysis," *Fire Safety Journal*, Volume 13, page 137-154, 1988.

[IAEA 1998]   **ISBN 92-0-103798-8**, "The Radiological Accident in the Reprocessing Plant at TOMSK," IAEA, 1998.

[IAEA 2008]   **IAEA**, "Safety of Reprocessing Facilities" (draft), October 2008.

[IAEA 2005]   **IAEA-TECDOC-1467**, "Status and Trends in Spent Fuel Reprocessing," IAEA, September 2005.

[IAEA 2007]   **IAEA-TECDOC-1529**, "Management of Reprocessed Uranium – Current Status and Future Prospects," February 2007.

[Ikeda 2003]   H. Ikeda, "Safety Design of Purex Liquid System – The Flash and Fire Points," *Journal of Nuclear Science and Technology*, Vol. 41 No.4, page 534-536, April 2004.

[INRA 2008]   CFTC Conceptual Design Studies, Final Report, Areva Report Number RPT 3000285, April 11, 2008.

[Iqbal 2004]   **NUREG-1805**, N. Iqbal and M.H. Salley, Fire Dynamics Tools (FDT): Quantitative Fire Hazard Analysis Methods for the U.S. Nuclear Regulatory Commission Fire Protection Inspection Program – Final report, U.S. Nuclear Regulatory Commission, Washington, DC, December 2004.

[IRSN 2008]   Technical Note – Risk of Explosion Associated with "Red Oils" in Reprocessing Plants, Institut De Radioprotection Et De Surete Nucleaire (IRSN), June 2008.

[Ishmatov 2010]   A.N. Ishmatov and B.I. Vorozhtsov, "Study of the Evolution of a Finely Dispersed Liquid Aerosol, Taking into Account the Effect of Hardly Evaporable Impurity," *Bulletin of the Lebedev Physics Institute*, Vol.37, No.1, pp.13-15, 2010.

[Jain 1998a]   V. Jain and R.T. Pabalan, "Review of Process Safety Issues Relevant to Vitrification of Radioactive Wastes," Proceedings of Waste Management '98, Waste Management Symposia, Inc., Tucson, Arizona, 1998.

[Jain 1998b]   **CNWRA 98-005**, V. Jain, "Survey of Solidification Process Technologies," Center for Nuclear Waste Regulatory Analysis, San Antonio, Texas, April 1998.

[Jordan 2003]   H. Jordan and M.T. Leonard, "Attenuation of Airborne Source Terms in Leak Paths," U.S. Department of Energy, LA-UR-03-7945, October 2003.

[Kerdrinskii 1993]   V.K. Kedrinskii, "Nonlinear Problems of Cavitation Breakdown of Liquids under Explosive Loading (review)," Translated from Prikladnaya Mekhanika I Tekhnicheskaya Fizika, Vol. 34 No. 3, pp. 74-91, May-June 1993.

[Kudryashova 2011]   O.B. Kudryashova et al., "Physicomathematical Modeling of Explosive Dispersion of Liquid and Powders," Propellants Explosive Pyrotechnics, 36 524-529, 2011.

[Lajitha 2008]   G. Lajtha, E. Raimond, and P. Kostka, "Severe Accident Codes for L2 PSA (ASTEC Requirements)," ERMSAR Meeting, Bulgaria, September 1, 2008.

[Liu 2003a]   G. R. Liu, "Mesh Free Methods: Moving Beyond the Finite Element Method," CRC Press, 2003.

[Liu 2003b]   M. B. Liu, G. R. Liu, K. Y. Lam, and Z. Zong, "Smoothed particle hydrodynamics for numerical simulation of underwater explosion," *Computational Mechanics*, 30, 106-118, 2003.

[Louie 2004]	D.L.Y. Louie and L.F. Restrepo, "Analytical Approach for Gas Explosion for DOE Nuclear and Non-Nuclear Facilities," OMICRON Safety and Risk Technologies, Inc., EFCOG SAWG 2004 Paper, 2004.
[Louie 2005]	D.L.Y. Louie and L.F. Restrepo, "Explosion Modeling for Accident Analysis Applications," OMICRON Safety and Risk Technologies, Inc., EFCOG SAWG 2005, May 3, 2005.
[Lu 2010]	X. X. Lu, L. Li, X. B. Ren, X. F. Yan, and Y. C. Dong, "Numerical simulations of interactions between shock wave and gas-liquid-air interfaces," The Third International Symposium on Bifurcations and Instabilities in Fluid Dynamics, *Journal of Physics*: Conference Series 216, 012012, 2010.
[Lucy 1977]	L. B. Lucy, "A numerical approach to the testing of the fission hypothesis," *The Astronomical Journal*, 82, Number 12, 1013-1024, December 1977.
[Manhattan 1999]	**DOE-MA-0001-01/99**, F.G. Gosling, "The Manhattan Project – Making the Atomic Bomb," US Department of Energy, 1999.
[Marsden 2010]	D. Marsden, et.al, "Vitrification Assistance Program: International Cooperation on Vitrification Technology (Part II) – 10157," Proceedings of Waste Management 2010, Waste Management Symposia, Inc., Tucson, Arizona, 2010.
[McLaughlin 2000]	**LA-13638**, T.P. McLaughlin et al., "A Review of Criticality Accidents (2000 Revision)," Los Alamos National Laboratories, May 2000.
[MELCOR 2004]	**U. S. Department of Energy**, MELCOR Computer Code Application Guidance for Leak Path Factor in Documented Safety Analysis, Office of Environment, Safety and Health, May 2004.
[Meess 2011]	D. Meess, "Vitrification of High-Level Radioactive Waste at the WVDP," West Valley Demonstration Project, WVES LLC, Presented at NWTRB Meeting in Buffalo, NY, April 27, 2011.
[Merrifield 1990]	R. Merrifield, "Report on the Peterborough - Explosion Blast Damage and Injuries," H M Explosives Inspectorate, UK, August 1990.
[MSF 2011]	Managing Spent Fuel from Nuclear Power Reactors – Experience and Lessons from Around the World, International Panel on Fissile Materials, 2011.
[Monaghan 1988]	J. J. Monaghan, "An Introduction to SPH," *Computer Physics Communications*, 48, 89-96, 1988.
[Monaghan 2005]	J. J. Monaghan, "Smoothed Particle Hydrodynamics." *Rep. Prog. Phys.* 68, 1703-1759, 2005.
[Mudali 1993]	U.K. Mudali et al., "Corrosion Studies on Materials of Construction for Spent Nuclear Fuel Reprocessing Plant Equipment," *Journal of Nuclear Materials*, 203, page 73-82, 1993.

[NEA 2012-15]   **Nuclear Energy Agency**, "Spent Nuclear Fuel Reprocessing Flowsheet – A Report by the WPFC Expect Group on Chemical Partitioning of the NEA Nuclear Science Committee," NEA/NSC/WPFC/DOC(2012)15, 2012.

[NFPA 2008]   **NFPA**, The SFPE Handbook of Fire Protection Engineering, 4th Edition, 2008.

[NRC 1974a]   **Regulatory Guide 3.18**, "Confinement Barriers and Systems for Fuel Reprocessing Plants," U.S. Regulatory Commission, Washington, DC, February 1974.

[NRC 1975a]   **Regulatory Guide 3.32**, "General Design Guide for Ventilation Systems for Fuel Reprocessing Plants," U.S. Nuclear Regulatory Commission, Washington, DC, September 1975.

[NRC 1975b]   **Regulatory Guide 3.31**, Revision O-R, "Emergency Water Supply systems for Fuel Reprocessing Plants," U.S. Nuclear Regulatory Commission, Washington, DC, September 1975.

[NRC 1976]   **Regulatory Guide 3.38**, "General Fire Protection Guide for Fuel Reprocessing Plants," U.S. Nuclear Regulatory Commission, Washington, DC, June 1976.

[NRC 1979]   **Regulatory Guide 8.21**, Revision 1 "Health Physics Surveys for By-Product Material at NRC-Licensed Processing and Manufacturing Plants," U.S. Nuclear Regulatory Commission, Washington, DC, October 1979.

[NRC 2010]   "Draft Regulatory Basis for a Potential Rulemaking on spent Nuclear Fuel Reprocessing Facilities," www.nrc.gov/materials/gap-summary-nrc-2010-0267-0004.pdf.

[NRC 1977]   **NUREG-0278**, K.J. Scheider et al., "Technology, Safety, and Costs of Decommissioning a Reference Nuclear Fuel Reprocessing Plant," Volumes 1 and 2, U.S. Nuclear Regulatory Commission, Washington, DC, October 1977.

[NRC 1988]   **NUREG-1320**, J.E. Ayer et al., Nuclear Fuel Cycle Facility Accident Analysis Handbook, U.S. Nuclear Regulatory Commission, Washington, DC, May 1988.

[NRC 2005a]   **NUREG-1767**, "Environmental Impact Statement on the Construction and Operation of a Proposed Mixed Oxide Fuel Fabrication Facility at the Savannah River Site, South Carolina," Final Report, Vol. 1, U.S. Nuclear Regulatory Commission, Washington, DC, January 2005.

[NRC 2005b]   "Final Safety Evaluation Report on the Construction Authorization Request for the Mixed Oxide Fuel Fabrication Facility at the Savannah River Site, South Carolina," 2005.

[Ogata 2003] S. Ogata, et.al, "Fluorination Reaction of Uranium Dioxide by Fluorine," *Journal of Nuclear Science and Technology*, Vol.41, No.2, page 135-141, February 2004.

[OECD 2005] **NEA No. 3588**, "The Safety of the Nuclear Fuel Cycle," Third Edition, Organization for Economic Co-Operation and Development, Nuclear Energy Agency, 2005.

[O'rourke 1987] P. J. O'Rourke and A.A. Amsden, "The TAB Method for Numerical Calculation of Spray Droplet Break-up," SAE Technical Paper 870289, (1987).

[Pace 2006] **INL/EXT-06-11969**, B. Pace, J. Braun, and H. Gilbert, "Idaho National Laboratory Fuel Reprocessing Complex Historic American Engineering Record Report – ID-3-H," Idaho National Laboratory, December 2006.

[Paddleford 1994] D.F. Paddleford and H.K. Fauske, "Safe Venting of Red Oil Runaway Reactions," WSRC-MS-94-0649, December 21, 1994.

[Paducah 2011] History of Paducah Gaseous Diffusion Plants, http://www.usec.com/gaseousdiffusion_pad_history.htm.

[Parker 1980] G.B. Parker, "Tributyl Phosphate Removal from Reprocessing Off-Gas Streams Using a Selected Sorbent," 16th DOE Nuclear Air Cleaning Conference, October 21, 1980.

[Perkins, 1980] **Savannah River Laboratory**, W.C. Perkins et al., "Potential Safety-Related Incidents with Possible Applicability to a Nuclear Fuel Reprocessing Plant," DP-1558, December 1980.

[Rodriguez 1995] E.A. Rodriguez and R.F. Davidson, "TOMSK-7 Radiochemical Facility Explosion: chemical Vessel Burst Failure Analysis," Report No. LA-UR-95-1756, Los Alamos National Laboratory, 1995.

[Schmidt 2010] **SAND 2010-0878**, R. Schmidt, et al., Foundational Development of an Advanced Nuclear Reactor Integrated Safety Code, Sandia National Laboratories, February 2010.

[Savannah50 Ch11] Savannah River Site at Fifty - Chapter 11, http://www.srs.gov/general/about/50anniv/50anniv.htm.

[Scheider 2001] M. Schneider et al., "Possible Toxic Effects from the Nuclear Reprocessing Plants at Sellafield (UK) and Cap De La Hague (France), Scientific and Technological Options Assessment," EP/IV/STOA/2000/17/01, European Parliament, November 2001.

[Scheider 2008] M. Schneider and Y. Marignac, "Spent Nuclear Fuel Reprocessing in France, A Research Report of the International Panel," April 2008.

[Shleien 1998] B. Shleien et al., Handbook of Health Physics and Radiological Health, 3rd Edition, Williams & Wilkins 1998.

[Standring 2006]   W. Standring, "Review of the Current Status and Operations at Mayak Production Association," StralevernRapport 2006:19, Norwegian Radiation Protection Authority, November 24, 2006.

[Steindler 1980]   M.J. Steindler and W.B. Seefeldt, "A Method for Estimating the Challenge to an Air Cleaning System Resulting from an Accidental Explosive Event," 16th DOE Nuclear Air Cleaning Conference, 1980.

[Sutter 1983]   **NUREG/CR-3093 (PNL-4566)**, S.L. Sutter, "Aerosols Generated by Releases of Pressurized Powders and Solutions in Static Air," prepared by Pacific Northwest Laboratory, U.S. Nuclear Regulatory Commission, Washington, DC, August 1983.

[Tanaka 1983]   **U.S. Department of Commerce**, T. Tanaka, "A Model of MultiRoom Fire Spread", NBSIR 83-2718, August 1983.

[Todd 2008]   T. Todd, "Spent Nuclear Fuel Reprocessing," Nuclear Regulatory Commission Seminar, Slide materials, Idaho National Laboratory, March 25, 2008.

[Ueda 2005]   Y. Ueda, "Current Studies on Utilization of Risk Information for Fuel Cycle Facilities in Japan," Workshop on Utilization of Risk Information for Nuclear Safety Regulation, Tokyo, May 10-11, 2005.

[Westinghouse 2011]   **Westinghouse**, FATE™ Software for Facility Process, Safety, and Decommissioning and Dismantling, NS-ES-0194, August 2011.

APPENDIX A

BARNWELL NUCLEAR FUEL PLANT DESCRIPTION

This appendix describes the Barnwell Nuclear Fuel Plant (BNFP) in terms of the major operations, designs, and processed data [NUREG-0278, AGNS 1975]. Major portions of BNFP have been constructed, which includes [NUREG-0278]:

1.   Main process building (26 m high, 36 m wide and 68 m long)

2.   Fuel Receiving and Storage Station (FRSS) (28 m high, 45 m wide and 45 m long)

3.   Control Room Area (CRA)/Plutonium Nitrate Storage and Load-out (PNSL) (18 m high, 20 m wide and 25 m long)

4.   Waste Tank Equipment Gallery (WTEG) (12 m high, 31 m wide and 34 m long) Waste Tank Cells (WTCs)

The plot plan for the BNFP is shown in Figure A-1. As shown in this plot plan, the overall process building including the FRSS and CRA and PNSL is shown in the center near the road entry at the south. Both WTSG and WTC are shown just west of the overall process building. The overall process building layouts are shown in Figure A-2 through Figure A-7. Shown in these figures are the wall thickness and dimensions of the building. In principle, thick concrete walls (up to 6 feet) are used in the area where high radiation is expected. For fuel storage pools, depth of water is used to guard against radiation. Windows for viewing at high radiation areas are constructed with high-density lead glass oil-filled materials to minimize any exposure. The lower right corner of the layout of the top view in Figure A-2 to Figure A-5 shows the FRSS. The upper right corner of the layout of the top view in Figure A-2 to Figure A-5 shows the CRA/PNSL and offices and cold/hot laboratories.

The throughput capacity of the BNFP was designed for reprocessing 5 metric tons per day using a PUREX separation process, which amounts to 1500 metric tons of spent fuel per year [AGNS 1975]. This plant can handle fuel with initial fissile content of up to 5% enrichment, or the equivalent for plutonium recycle fuels.

The overall detailed process is presented in Figure A-8. In Figure A-9, a simplified process block diagram is presented. As shown in this figure, the spent fuel is received in heavily shielded casks and is unloaded and stored underwater in a pool in the FRSS. During processing, each fuel assembly is transferred under water to the main process building, where fuel assemblies are partly disassembled, chopped into pieces up to 5" long and dropped into a dissolver vessel. The dissolver vessel contains sufficient nitric acid to dissolve the fuel. Volatile fission products (such as noble gases, tritium halogens and carbon) are released to the effluent cleanup system during this step. The fuel cladding hulls and assembly parts are packaged and taken to a bunker-type interim storage area outside.

The dissolved fuel material (in nitric acid aqueous solution) is centrifuged and sent to the first stage of solvent extraction using organic solution of 30% tributyl phosphate (TBP), where the uranium and plutonium are extracted into the organic phase in a centrifugal contactor while most of the other transuranic elements and fission products remain in the aqueous nitrate solution. This nitrate solution also contains about 0.5% of the unrecovered uranium and

plutonium, which goes into the high-level liquid waste (HLLW). After concentration, the HLLW is transferred to underground tanks for interim storage.

The organic stream (plutonium and uranium) from the contactor is scrubbed in a pulse column contactor to remove additional fission products and other transuranic elements. Then this solution is processed through an electro-pulse column where the chemical valence of the plutonium is reduced, so that the uranium can be separated. The plutonium aqueous stream is processed through two additional stages of solvent extraction to remove residual fission products and other impurities. Uranium in the organic solvent solution is processed through a solvent extraction to remove any impurities. Silica gel filtration adsorption is used in the final uranium solutions to remove any residual zirconium. The uranium nitrate conversion to the uranium hexafluoride facility is included in the BNFP. However, the plutonium nitrate is stored in tanks and sent to other facilities for further processing. The waste streams at the BNFP include intermediate and high level wastes, which are stored in tanks, waiting for further solidification and disposal. The nitric acid used in the various processes is recovered for re-use. All aqueous raffinates containing small quantities of fissile material, except solvent treatment wastes, are passed through a recovery extraction system prior to concentration and storage. Iodine scrubbers and inorganic iodine adsorption beds are used to minimize any iodine effluent through the stack. The iodine remaining in the dissolver solution may be removed by mercury addition to the general purpose concentrator and acid fractionator overhead vaporizer before discharge to the stack. The high level wastes are chemically denitrated to a nitric acid concentration of 1-5 Molar before being sent to interim liquid waste storage. Details of the processes at BNFP will be described in the following sections.

In this appendix, the descriptions of each of the above 5 areas of the BNFP is discussed along with the process operations. Support systems and operations are also described, especially safety related.

Chemical inventory at the BNFP includes nitric acid, gadolinium nitrate, tri-butyl phosphate, hydrazine, hydrogylamine and mercury [NUREG-0278, vol.1 5-27].
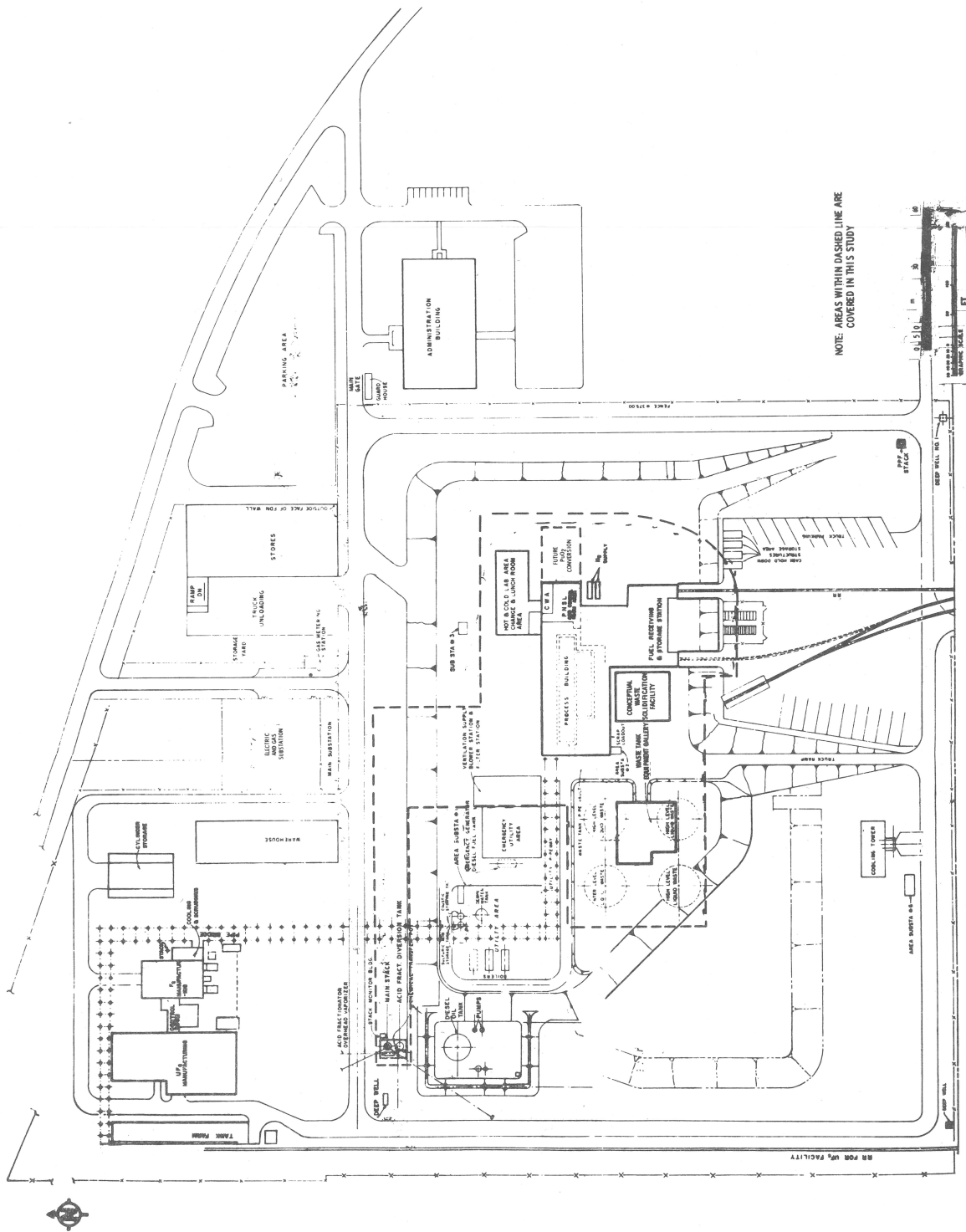
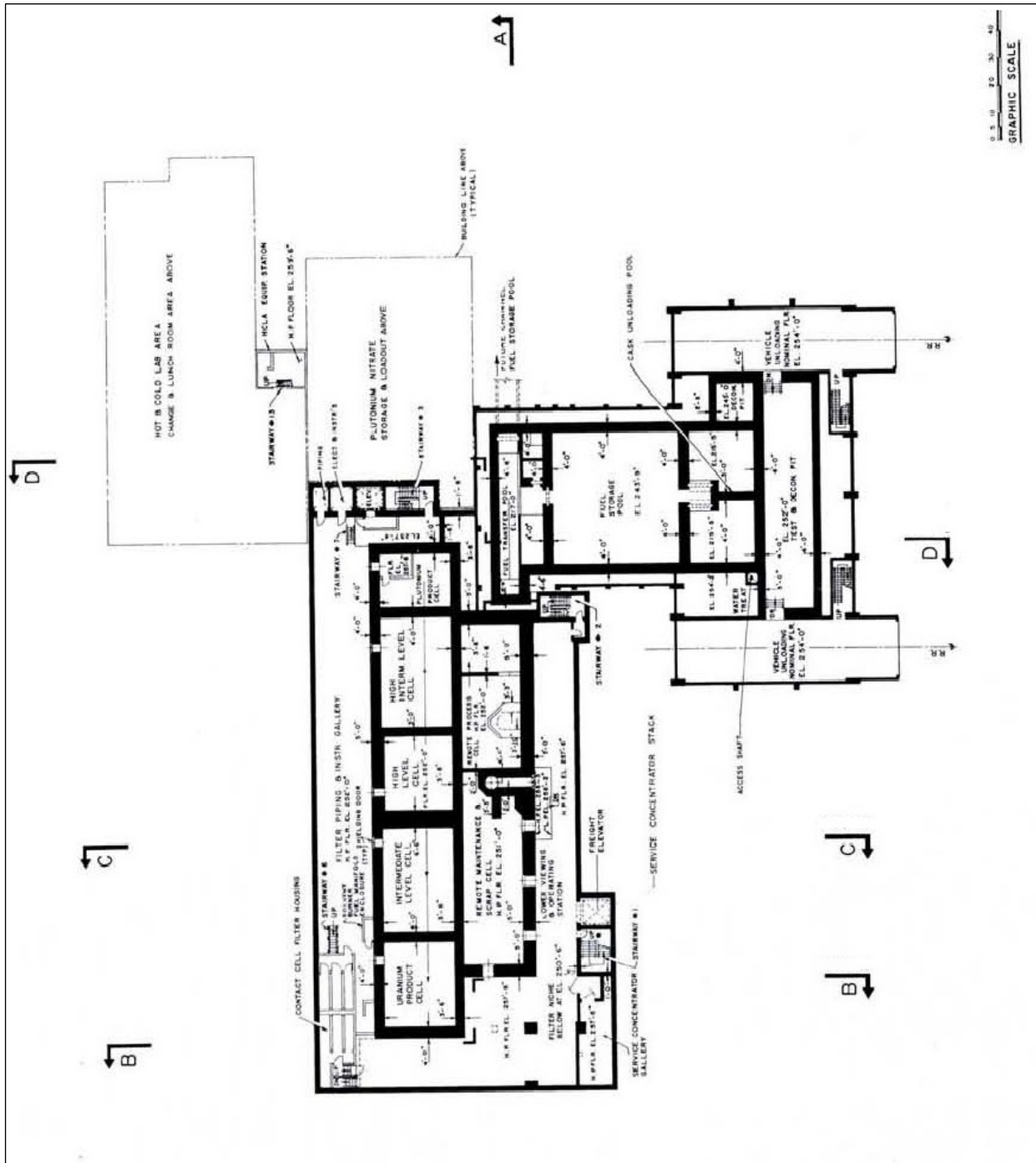**Figure A-1   Plot Plan for BNFP [NUREG-0278]**

Figure A-2   Top view of overall process buildings at elevation 270' 6" [AGNS 1975]

Figure A-3   Top view of overall process buildings at elevations 270'-6" to 279'-0" [AGNS 1975]

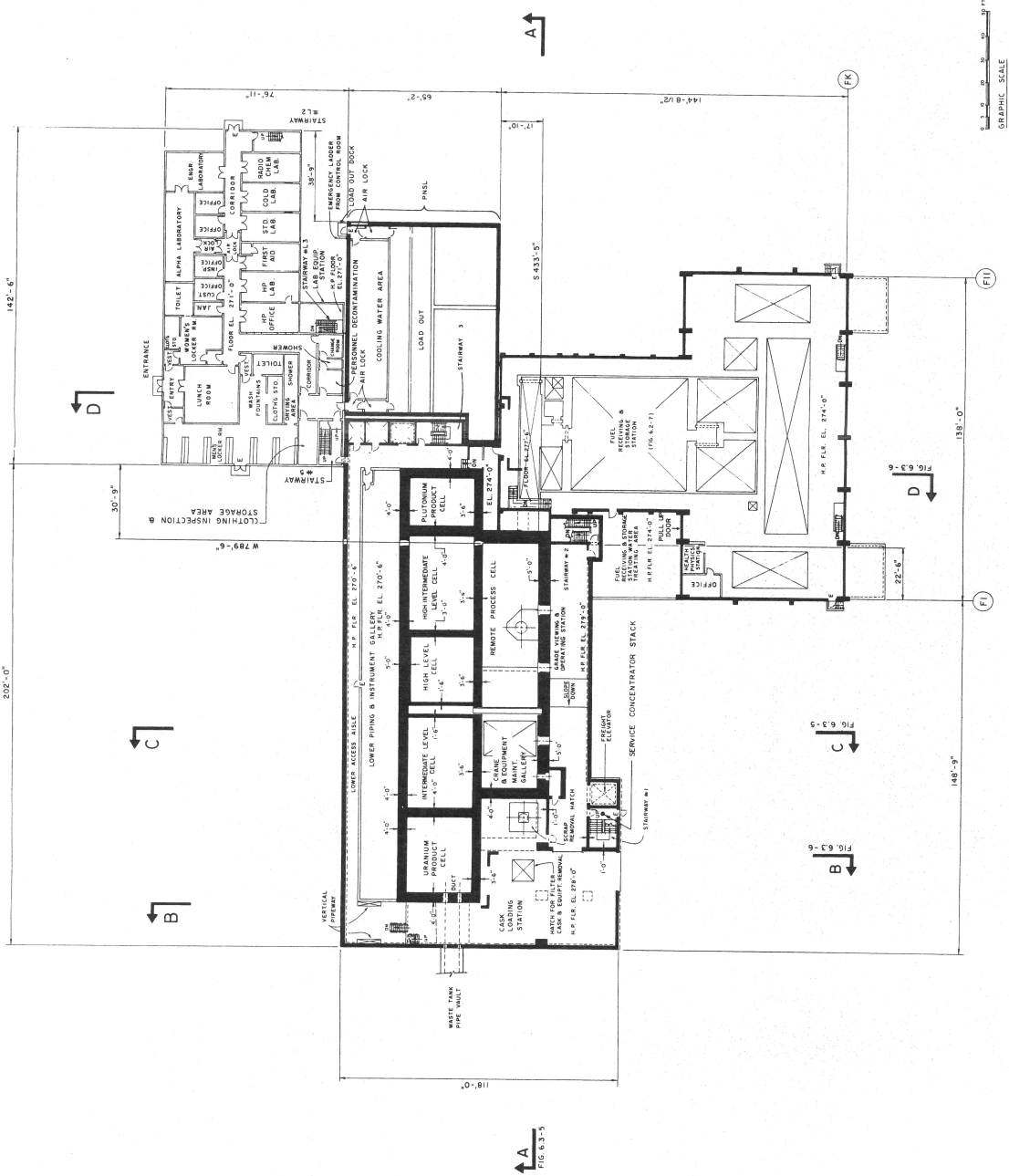**Figure A-4   Top view of overall process buildings at elevations 279'-0" to 297'-0" [AGNS 1975]**

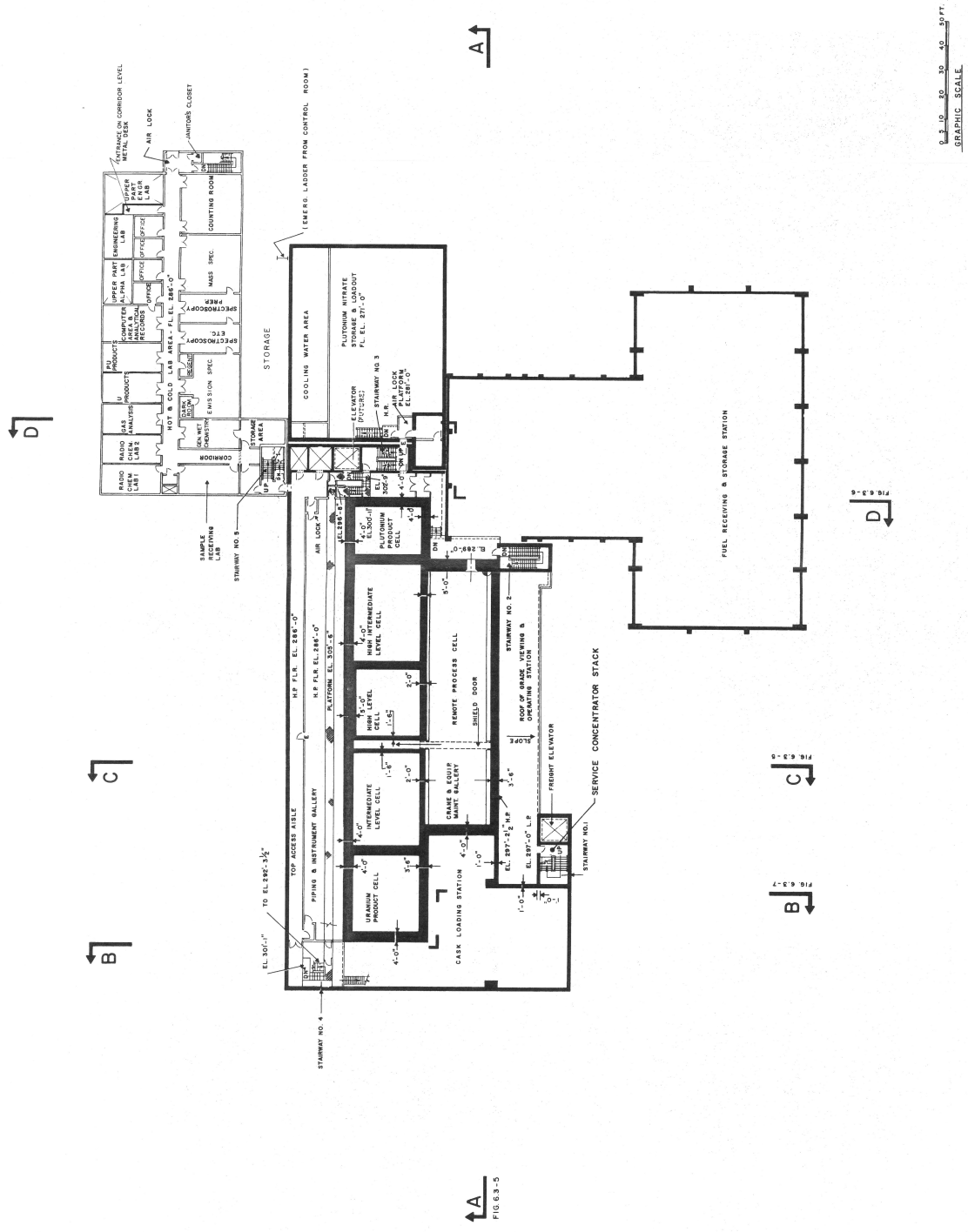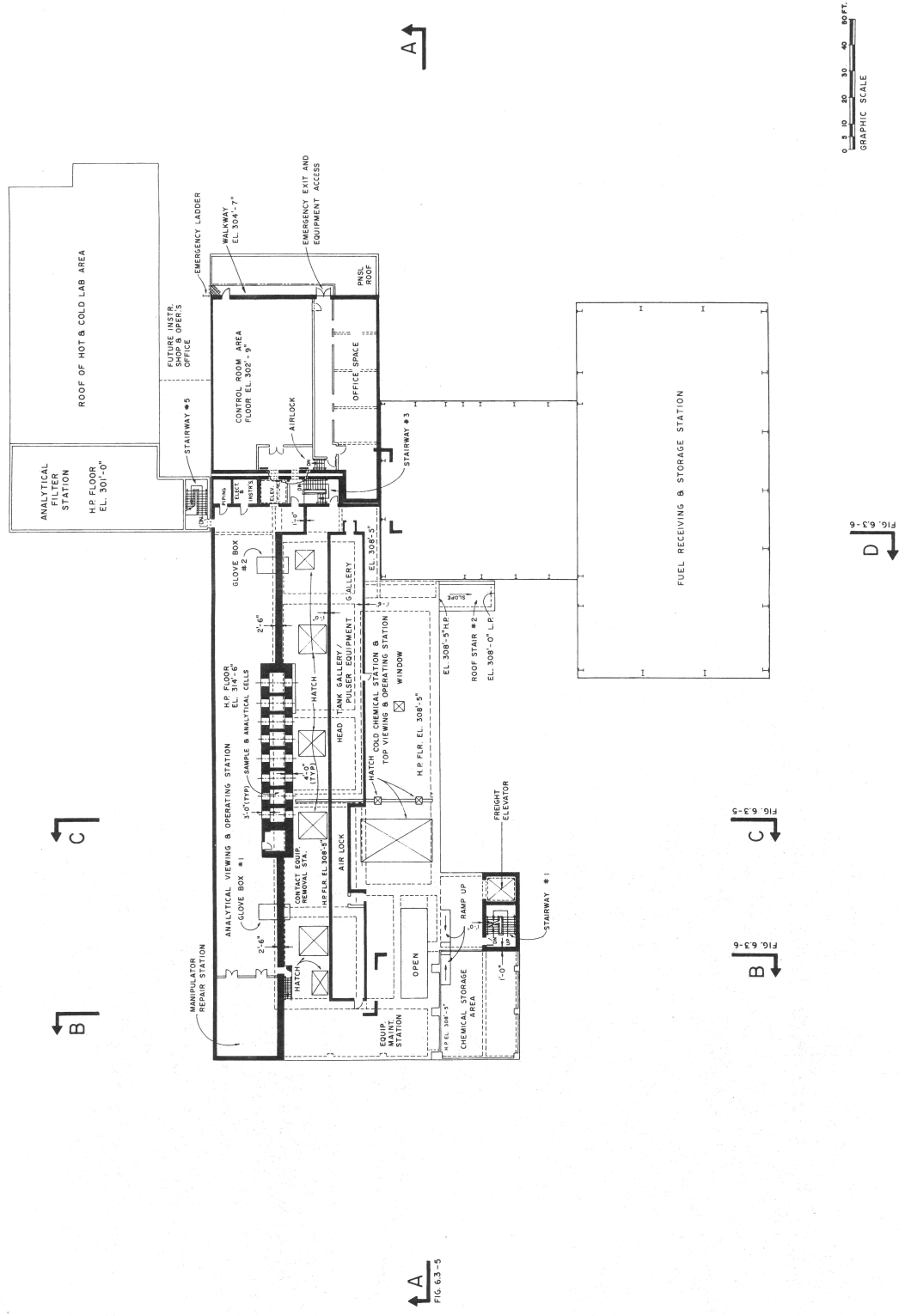**Figure A-5   Top view of overall process buildings at elevation above 297'-0" [AGNS 1975]**
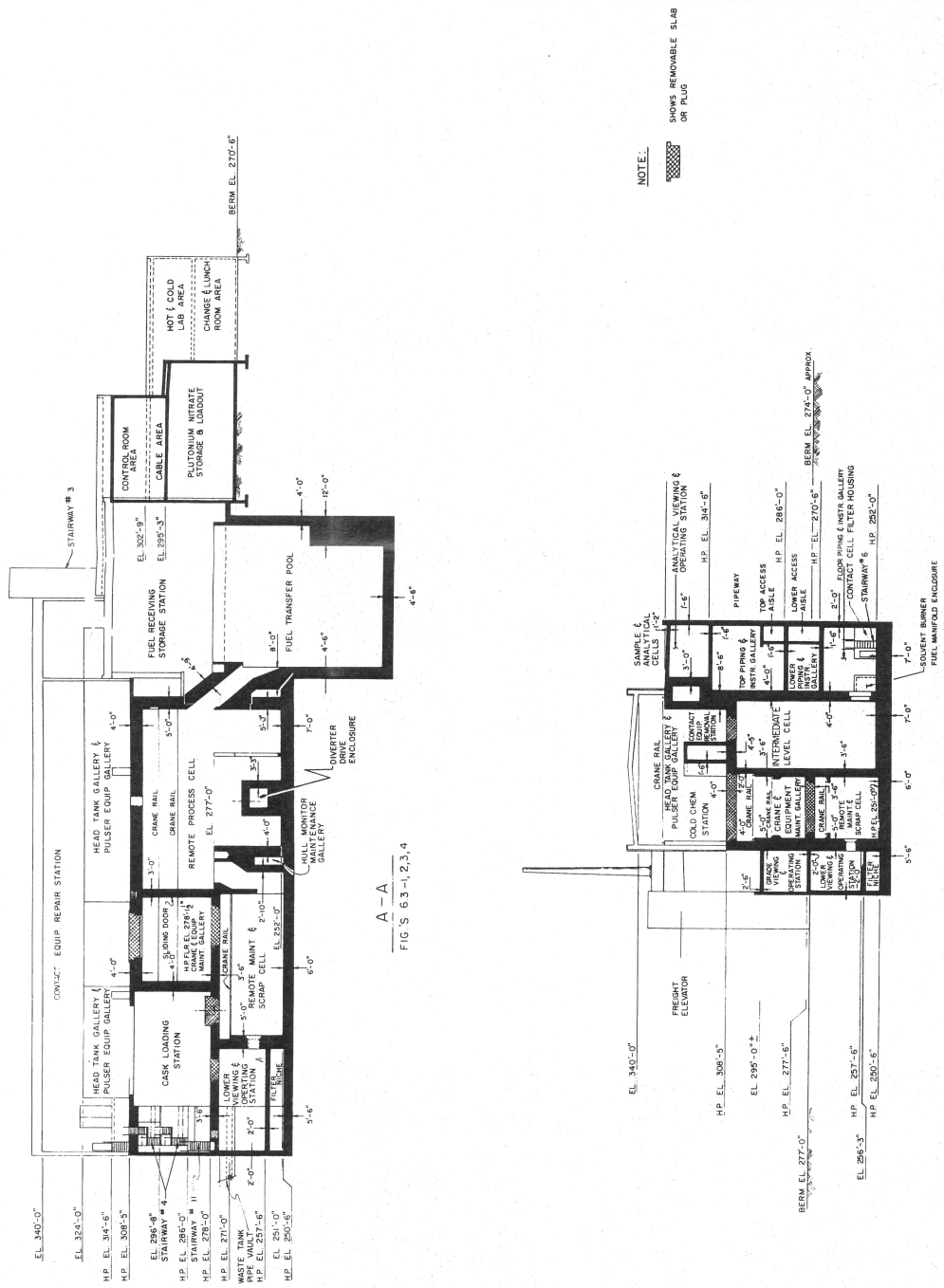
Figure A-6   Side view of overall process buildings at A-A and C-C planes [AGNS 1975]
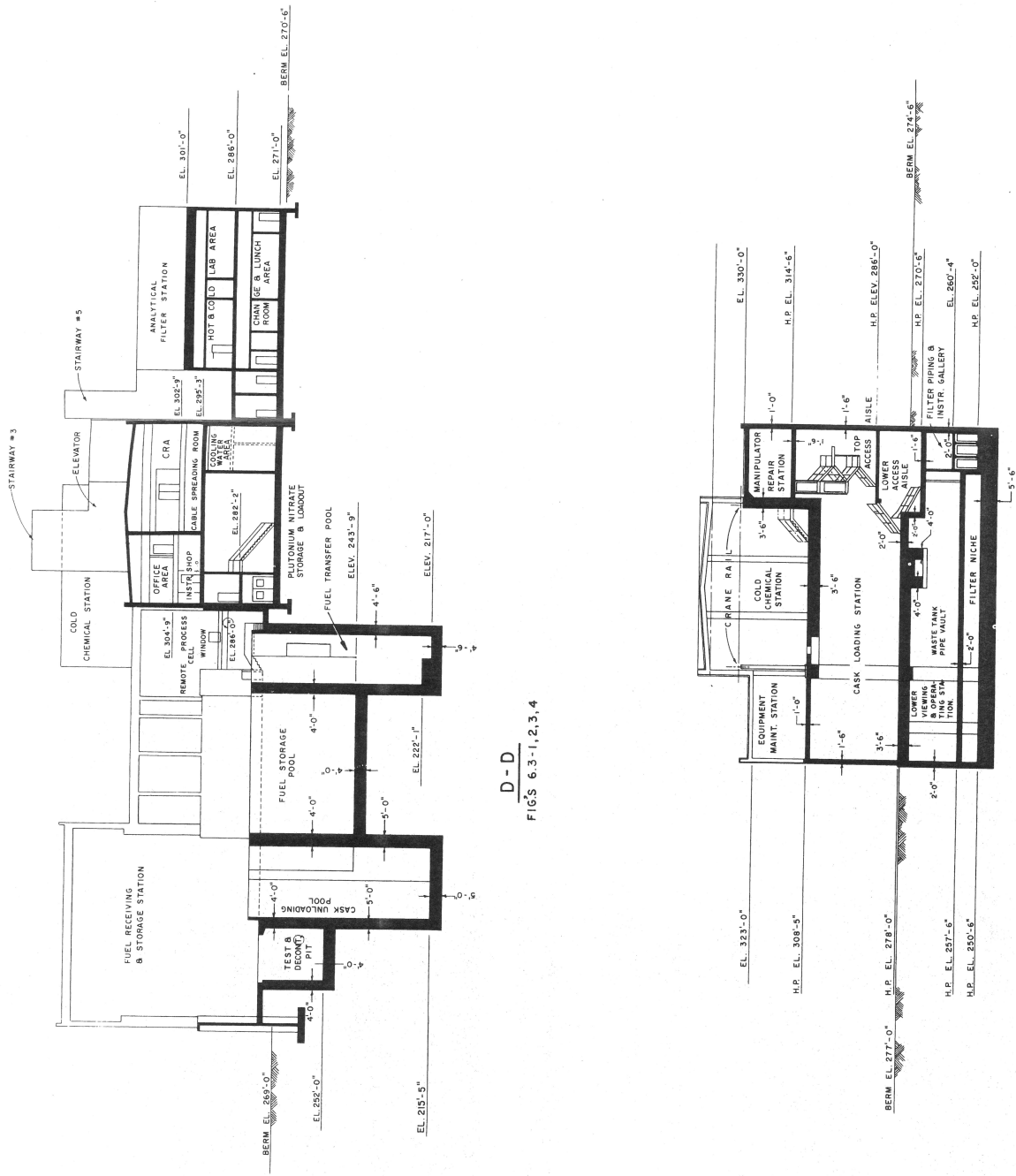
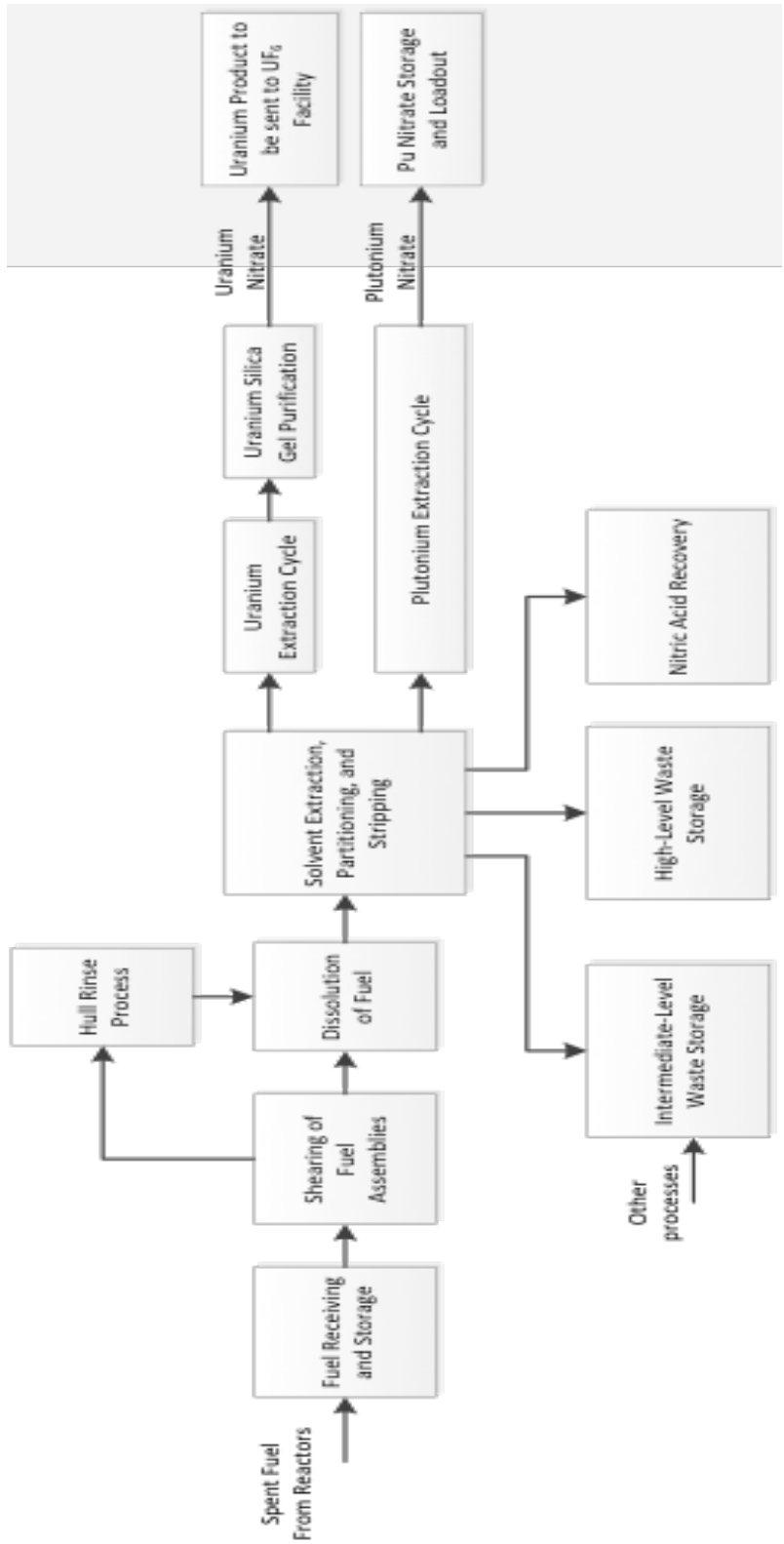**Figure A-7   Side view overall process buildings at D-D and B-B Planes [AGNS 1975]**

Figure A-8   Overall Process Diagram for the BNFP [AGNS 1975]

Figure A-9   Simplified block process flow diagram

A.1   Main Process Building

The main process building contains the majority of the processes at the BNFP, which include:

1.   Shearing of fuel assemblies
2.   Dissolution of fuel
3.   Solvent extraction, partitioning and stripping
4.   Fissile extraction cycle (uranium and plutonium)

Structures included in this building are the following (see Figure A-2 to Figure A-7):

1.   Remote Process Cell (RPC)
2.   Remote Maintenance and Scrap Cell (RMSC)
3.   High Level Cell (HLC)
4.   High-Intermediate Level Cell (HILC)
5.   Intermediate Level Cell (ILC)
6.   Uranium Product Cell (UPC)
7.   Plutonium Product Cell (PPC)

The following sections describe each of the structures in more details, including the operations within each structure.

A.1.1   Remote Process Cell (RPC)

This cell is a high-level radiation area used primarily for the head-end mechanical process operations and initial chemical processing. As its name implied, all operations conducted in this cell are performed remotely. The fuel assemblies are transferred from the fuel transfer pool of FRSS to the feed mechanism of the mechanical shear in the cell via the use of overhead crane through a transfer tunnel between this cell and FRSS (see Figure A-10). The mechanical shear chops the fuel assemblies into small segments about 2 to 5 inches long. These segments are contained in the dissolver basket inside the dissolver that contains hot nitric acid and gadolinium nitrate (which acts a soluble neutron absorber). The un-dissolved cladding (hulls of stainless steel and Zircaloy) remaining in the basket is lifted from the dissolver and is transferred to RMSC.

Figure A-10   A-A View of the equipment arrangement in remote process cell [NUREG-0278]

During the chop and shear process, the sweep-air system provides a positive flow of air from the cell into the shear feed magazine, so that the air sweeps any fission product gas and particulate down the diverter chute into the dissolver system to prevent any release into the cell during shearing.

The cell floor and lower walls are lined with 304L stainless steel to contain spills of the process solutions. The stainless steel liner is welded to anchors in the underlying concrete. The spilled solutions may be transferred from the cell sumps to the intermediate level liquid waste system or returned to the process. Any exposed concrete surfaces in the cell are coated with an acid-resistant phenolic paint system such as Amercoat 90 or Phenoline 305. Most of the external walls contain 5' thick concrete where the high radiation is expected. The 5-foot thick concrete structure provides shielding to personnel in normal operating areas surrounding the RPC to a radiation exposure no more than 1 mrem per hour. Interior walls and lower radiation area (above the process equipment) are generally thinner. There is a removable (wheel-mounted) shield door allowing the crane and power manipulator access to this cell from the Crane and Equipment Maintenance Gallery (CEMG) where the equipment is normally stored and maintained (see Figure A-4). See Section A.6 for the detailed description of CEMG function. There are five RPC shielding windows and frames are designed to withstand an operating basis earthquake (OBE) and design basis earthquake (DBE) or an in-cell blast and still maintain confinement. Table A-1 lists the major equipment in RPC.

Table A-1 Major equipment description in RPC [AGNS 1975]

| Equipment	Operations	Descriptions
Sheared fuel diverter	Once the fuel assemblies are chopped, the fuel segments are diverted to the dissolver	This diverter has a dimension of 8'9" diameter × 11'1" high.
Fuel dissolver (15-C-101, 102, 103)	The purpose of the dissolver is to perform the following dissolution sequence (see Figure A-11 for the dissolver flow diagram): 1. Chopped fuel is placed into approximately 3-8 Molar HNO_3 and 3 g/liter gadolinium recirculating dissolver solution in the dissolver basket. 2. Dissolver product is withdrawn continuously by air lifting the feed surge tank when solution is within proper density of ~ 1.49 g/cc. 3. After the dissolve cycle, the dissolved fuel is exposed to simmer solution (8 Molar HNO_3 and 3 g/liter Gd), which is air lifted to the dissolver section for dissolving the incoming chopped fuel. 4. Leached hulls are rinsed with hull rinse solution (0.2 Molar HNO_3 and 7.5 g/liter Gd). 5. Leached hulls contained in the removable dissolver basket are dumped in the hull loadout station.	The dissolver includes three identical dissolver barrels, each capable of operating in dissolve, simmer, or rinse modes as previously described. Each right cylindrical barrel, which is made of 304L stainless steel, measures in 3'2" in diameter at the top section, 2'7.5" in diameter at the bottom, and 15'7" high. Each barrel is also accompanied by a heat exchanger (15-E-132, 133, 134) to maintain proper temperature. This exchanger contains a heating capability to 240 ˚F during simmer cycle and to cool to 190 ˚F during dissolve cycle. The process fluid is outside the shell. Heating or cooling uses neutron poisoned water on the tube side.
High activity waste (HAW) concentrator	This equipment concentrates the high-activity waste steam (which contains about 99% of the nonvolatile fission products from spent fuel) from the extraction operations (see Figure A-1). The concentrate is sampled and then sent to the high level liquid waste storage facilities.	This concentrator (41-E-460) is constructed of titanium (vessel and jacket). It uses a titanium thermosyphon reboiler that is supplied with 25 psig steam at 260 ˚F. It measures 5'3' in diameter × 18'0.25" high.
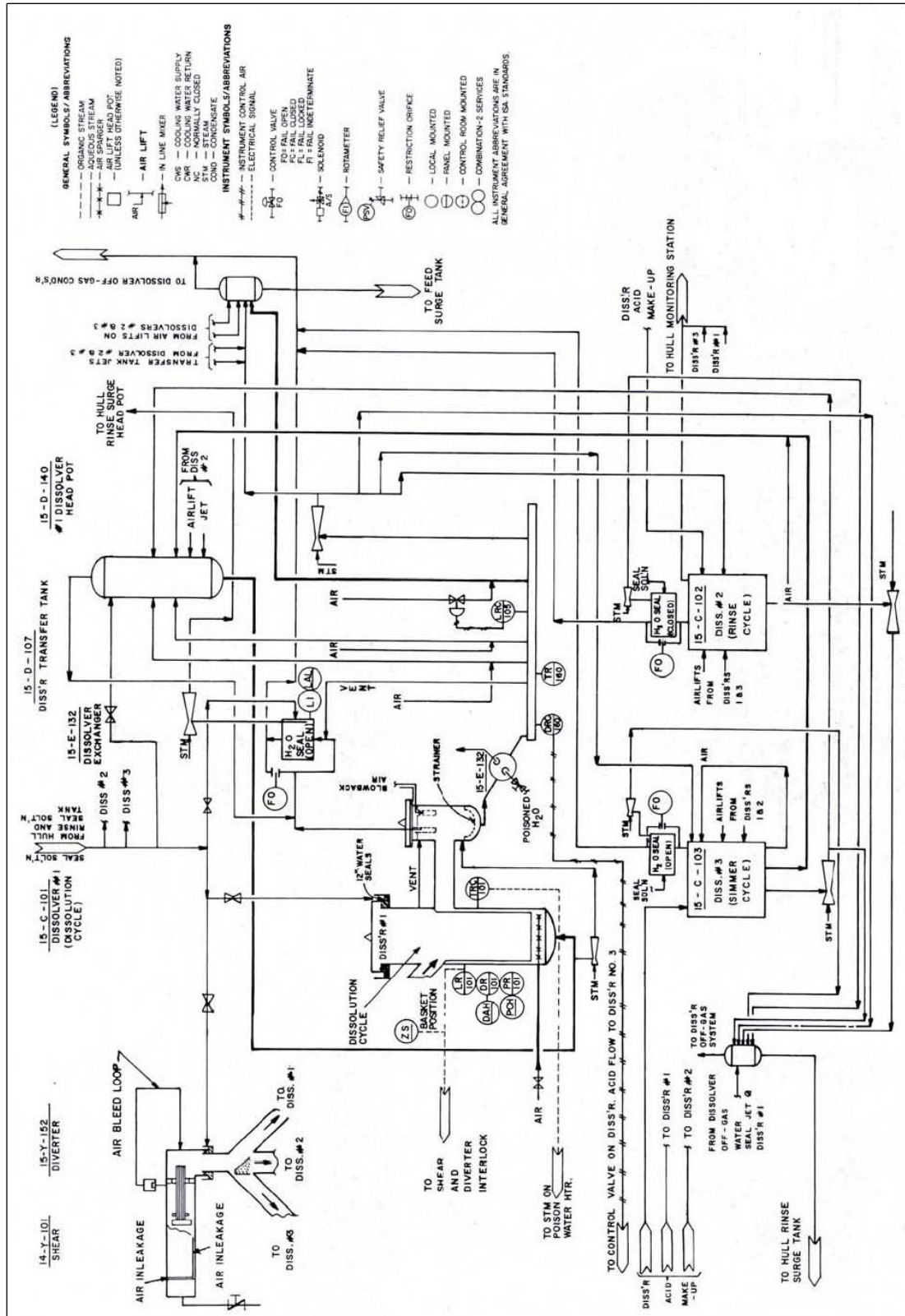
Figure A-11   Typical dissolver flow diagram [AGNS 1975]

A.1.2   Remote Maintenance and Scrap Cell (RMSC)

This cell is used to manage the scrap from the cladding hulls and other radioactive scrap. In addition, this cell is used to decontaminate and repair process equipment. As its name implied, all operations are done remotely. All walls and floor are lined with 5-mm thick 304L stainless steel. Exposed surfaces other than the stainless steel are coated with phenolic paint.
Figure A-12 shows the layout of the cell and its equipment. As shown in this figure, both A-A and B-B views show the locations of the dissolver basket dumper which is located at the roof line of the cell, the cladding hull disposal containers, and the 5-ton overhead crane used in the container transfer and for other purposes. When the filled dissolver basket is transferred from the remote process cell to this cell where the dumper is located, the dumper rotates the basket to allow all cladding scrap to fall into the chute which forms the lower portion of the dumper, where it is dropped into an open concrete hull disposal container (measuring 3' inside diameter and 7'4" high) with a total capacity of 52.5 ft^3. Sand can be added to the container after dumping, if the spent hull fines should become ignited (Zircaloy fines can be pyrophic under certain conditions). A total of three dissolver baskets full of the spent hull can be contained inside a disposal container. Once it is capped, it is moved into a storage location by a hull transfer car. The dumping station is located at one end of the cell and the loadout station at the other, where the Cask Loading Station is located.   The loadout station is used to load the cask with the decontaminated filled disposal containers at the cask loading station above the cell. A removable hatch separates this cell and the cask loading station. In addition, a direct straight-line 8-inch chute from the Sample and Analytical Cell (SAC) is provided to transfer solid waste to the SAC waste retainer in the RMSC. Other activities that can be performed in the cell include: examination of partially sheared fuel assembly via the fuel transfer hatch, and conduct large equipment maintenance, examination and decontamination from the remote process cell to this cell via a hatch located between the floor of the CEMG and the ceiling of this cell.

Figure A-12   Equipment arrangement in RMSC [NUREG-0278]

A.1.3   High Level Cell (HLC)

This cell is used to provide chemical adjustment and centrifuging of the main process stream from the dissolver solution of the remote process cell. It also handles the high and intermediate level waste solutions. The cell is designed and constructed (with reinforced concrete) to provide confinement during normal, abnormal operations, seismic or meteorological conditions. This cell is equipped with a controlled ventilation supply/exhaust system to confine gaseous discharges and to transfer such discharges to the process ventilation system for treatment, monitoring, and discharge. The doors, hatches and penetrations which are an integral part of the structure do not compromise its confinement and shielding functions. This cell is designed

for remote operation and contact maintenance. This cell contains a stainless steel floor pan and sump to catch any process spills. Exposed concrete surfaces are coated with phenolic paint. There are two ways to access this cell. The first one is through a shield door from the Filter Piping and Instrument Gallery (FPIG), and the second one is through a removable concrete ceiling hatch to the Contact Equipment Removal Station (CERS). This cell contains a fire detection and suppression system. In addition, the sumps located in this cell are configured to prevent an advertent criticality. This cell has a dimension of 26ft × 28ft × 51.5ft [AGNS 1975]. The major equipment in the cell is listed in Table A-2.

Table A-2 Major equipment descriptions at HLC [AGNS 1975]

Equipment	Operations	Descriptions
Hull rinse surge tank	It is used in the hull rinse process.	It has a dimension of 5' diameter × 8'1" high
Dissolver acid surge tank	It is used to contain the feed from the hull rinse surge tank and the recovered nitric acid feed before sending the solution back to the dissolver.	It has a dimension of 5'10" diameter × 9'10" high [NUREG-0278].
Dissolver head pot tank	It is a disengaging vessel for air lefts and steam jets.	A total of four such tanks exist in the cell. Each measures 1'1" diameter × 3'8" high [NUREG-0278]
Accountability tank	It provides input accountability point. Dissolver solution is mixed and then accurately measured for volume and density, and sampled for chemical analyses.	It measures 6'2" diameter and 14'9" high [NUREG-0278]
Feed adjustment tank	It provides surge and batch adjustment of acidity and concentration of feed to recovery section.	A total of 2 tanks are located in the cell. Each measures 6' diameter × 15'8" high [NUREG-0278] .
General purpose (GP) concentrator feed tank	It receives process wastes continuously and others on a batch basis. It also receives feed of oxalate solutions from the silica gel bed regeneration and low activity general process wastes. The oxalate is destroyed by periodically raising the solution temperature in the tank to boiling, and adding hydrogen peroxide.	It measures 8' diameter × 15'1" high [NUREG-0278].

A.1.4 High Intermediate Level Cell (HILC)

This cell is to provide the solvent extraction of uranium and plutonium from fission products, separation of uranium from plutonium, concentrating intermediate-level liquid wastes and solvent cleanup and storage. The high activity (HA) centrifugal contactor (see Figure A-13) and IB electro-pulse column are installed in removable shield plugs in the ceiling. Most of the process equipment in the cell is constructed of stainless steel. The IB electro-pulse column has titanium and platinum internals. Table A-3 lists the major equipment in the cell.

Under normal process conditions, uranium concentrations are maintained well below 75% of the minimum critical concentration corresponding to the specified maximum enrichment. In the event of DBE, major components including the connecting piping must withstand the accident condition. In addition, a sump located on the cell floor contains instrumentation for detection of the process losses. Two jet sump transfer systems are used to remove liquids from the sump: one system is designed to operate under all abnormal conditions to transfer the liquid to the HLLW tanks, and the other is designed for normal operating transfers to the sump collection tank. It has a dimension of 40ft × 27ft × 52ft [AGNS 1975].
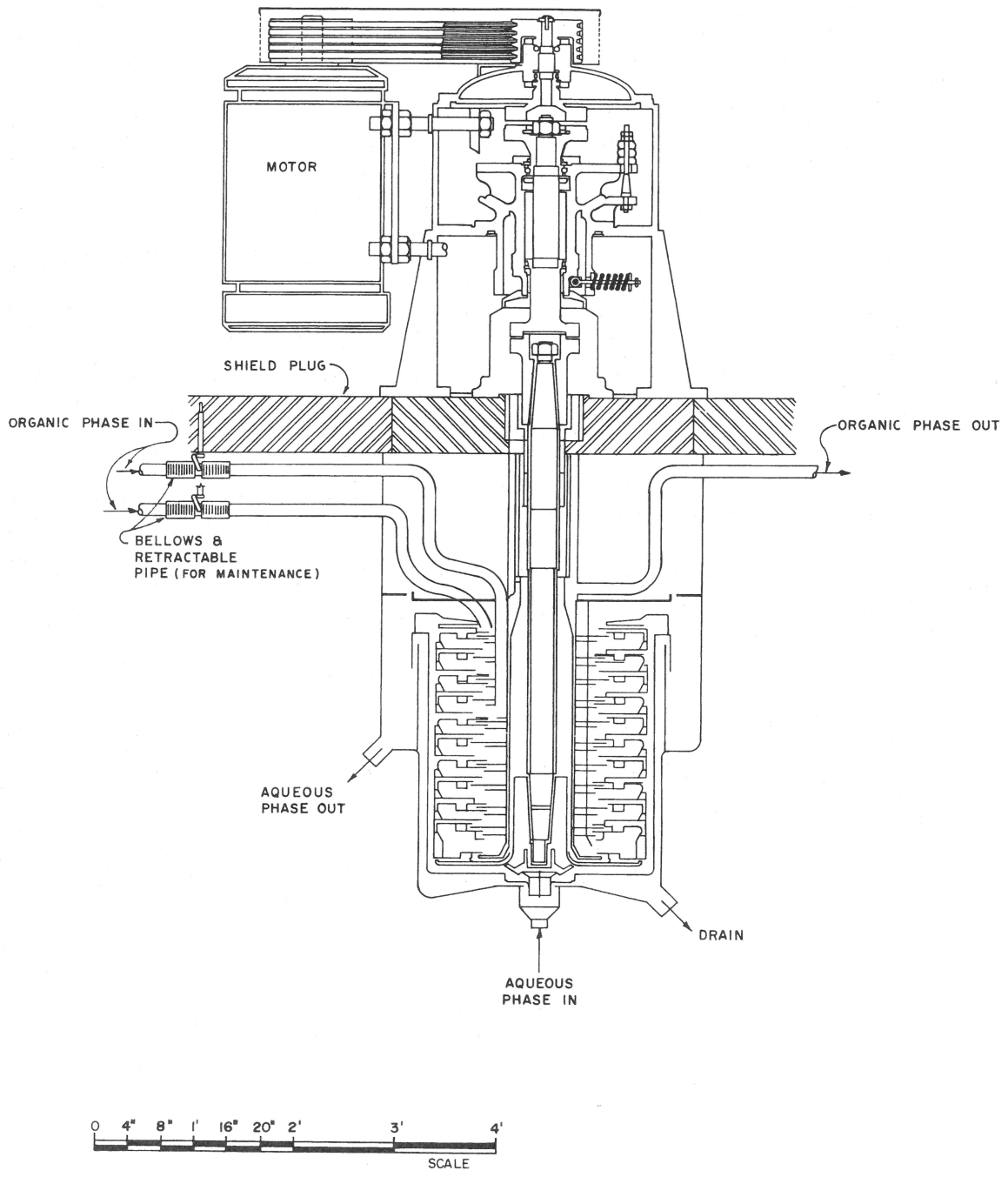
Figure A-13　HA centrifugal contactor in HILC [AGNS 1975]

Table A-3   Major equipment descriptions in HILC [AGNS 1975]

Equipment	Operations	Descriptions
Centrifugal contactor (21-C-225)	After acid adjustment, the feed (about 70% is the adjusted dissolver product from the HAF feed tank and about 30% is the HS column raffinate) is clarified by this equipment, and the solution is contacted counter-currently in 10-stage with an organic solution of 30% tributyl phosphate (TBP) in normal paraffin hydrocarbon diluent (dodecane). The organic solution preferentially extracts the tetravalent plutonium and hexavalent uranium, leaving about 95% of fission products in the aqueous solution. The aqueous solution is feed to the HAW surge tank in RPC. The organic solution is feed to HS column.	It is about 3'1" diameter × 4'9" high for the section below the shield plug (see Figure A-12). The contact time between phases is 10 minutes, compared to two hours in typical pulse columns. The reduction of contact time radically reduces solvent degradation due to radiation. The 10-stage centrifugal contactor used was developed and is used in France at the La Hague Plant.
HS column	The organic solvent stream from the centrifugal contactor is heated to 60 ˚C to promote back-extraction of additional fission products during the scrubbing action of a relatively high acid stream (3 Molar HNO_3).	It measures 1'10" diameter × 46'9" high [NUREG-0278]. This equipment has been met the criticality safety requirement, which is based on the fissile material concentration expected.
Electro-pulse column	This column is used to partition uranium and plutonium without the use of chemical reducing agents such as ferrous sulfamate. The concept is based on electrolytic reduction of U^{+6} to U^{+4}. The reduced U then reduces Pu^{+4} to Pu^{+3} so that the plutonium is extracted into the aqueous phase, effecting a separation of Pu and U. The Pu stream is transferred to the 1BP surge tank before sending to the PPC.	The plutonium concentrations in the electro-pulse column are maintained at approximately 75% of the minimum critical concentration. For the abnormal condition where feed to the column continues, but no plutonium is removed, the time required to achieve a critical concentration is conservatively estimated to be > 6 minutes. Alarm system is in place to ensure continuously flow of plutonium out of the column. In addition, hydrogen can be generated in this column under certain conditions. The column vent is continuously monitored to detect any potential explosive conditions.
General purpose (GP) concentrator	It receives feed from the GP concentrator feed tank from HLC by a continuous air lift. The concentrator liquid is processed to a specific gravity of 1.35, at which time the concentrate is batch transferred to GPW check tank.	It measures 7'8" diameter × 15' high [NUREG-0278]. Note that steam to the concentrator is supplied from the 25-lb closed loop system. This ensures that processing temperatures below 135 ˚C (minimum required for "red oil" explosion) are maintained. This concentrator is equipped with interlock systems that shut off the steam to the concentrator reboiler automatically in the event that pressure or temperature parameters are exceeded. In addition, sugar solution is added to the concentrator continuously to maintain a residual nitrite concentration and suppress ruthenium volatilization. Iodine in the feed may be removed by mercury addition before discharge to the stack.
GP waste check tank	It receives waste from the GP concentrator. It also receives mercury-bearing solutions from iodine scrubbers and purged solutions of the acid fractionator from the ILC. When it is full, the concentrate is sampled for	It measures 2'10" diameter and 6'8" high [NUREG-0278]. .

Table A-3   Major equipment descriptions in HILC [AGNS 1975]

Equipment	Operations	Descriptions
	accountability, H$^+$ concentration and activity. Then it is transferred to the intermediate level liquid waste tanks.	
1SF tank	It collects all aqueous process wastes, except those in the HAW stream. It feeds to the 1S column.	It measures 3' diameter × 31'7" high [NUREG-0278]. This tank is designed to safely contain higher plutonium concentrations than the 1S column, since this tank feeds to the 1S column. Measurement is done to monitor the contents. Fixed poison of Borosilicate glass raschig rings is added to the tank to minimize criticality potential.
1S column	It functions to recover fissile materials from wastes generated by 2nd uranium cycle, and 2nd and 3rd plutonium cycle. The fissile material is extracted as the organic phase via air lift from its top to the 1BU recycle tank. The remaining aqueous waste is air lifted to the LAW concentrator feed tank in ILC.	The main section is about 1' diameter × 25' high with a bottom disengaging section ~ 3' diameter × 3' high.
1BP Surge tank	It receives the Pu stream from the electro-pulse column.	It measures 4' diameter × 14'8" high. Borosilicate—glass Raschig rings are added to the vessel for criticality safety.

A.1.5   Intermediate Level Cell (ILC)

This cell houses the off-gas treatment, spent solvent treatment and combustion, acid recovery, and low-level liquid waste concentration. All operating activities are performed remotely. Most equipment and piping are constructed of 304L stainless steel; otherwise it is noted. This cell has a dimension of 27ft × 38ft × 52ft. The major equipment in the cell is listed in Table A-4.

Table A-4 Major equipment descriptions in ILC [AGNS 1975]

Equipment	Operations	Descriptions
NO$_2$ absorber	It is a dissolver off gas absorber to scrub any NO$_2$, primarily from the dissolvers.	It measures 5' diameter × 31'7" high.
LAW concentrator, feed tank, reboiler	It concentrates feed from the NO$_2$ absorber, and the acid stream from the HILC. Approximately the concentrate is about 20 fold than the aqueous waste from the feed tank. Once it is distilled, the overhead vapor containing about 3% nitric acid in water vapor is fed to the acid fractionator. The concentrate is sent to the LAWB check tank.	The concentrator measures 7'10" diameter × 20'8" high. The reboiler measures 4' diameter × 13'8" high. The feed tank measures 7'4" diameter × 16'6" high [NUREG-0278]. To ensure that the plutonium concentration is critical safe. Soluble poison may be added to the LAW concentrator feed tank based on the plutonium monitor upstream to the feed tank.
LAWB check tank	This tank samples the feed, then the feed is transferred to the HAW surge tank in HLC.	It is about 3' diameter ×6' high. This tank equipped with a sampler, a cooling water jacket and air sparger.
Acid fractionator, reboiler, and condenser	The fractionator is used to concentrate and recover nitric acid. The reboiler supplies heat to distill and fractionate dilute nitric acid to form	The fractionator measures 5'6" diameter and 38'8" high [NUREG-0278]. It is constructed of titanium. Mercury addition may remove

Table A-4 Major equipment descriptions in ILC [AGNS 1975]

Equipment	Operations	Descriptions
	12 Molar acid and water. In addition, the fractionator has draw offs to remove chlorides (nitrosyl chloride), thus preventing accumulation of corrosion contributing chlorides. Chloride concentrations above 0.05 wt% are corrosive. Therefore, purging is performed once a month to reduce this concentration below this limit. The purged solutions are sent to the GPW check tank in HILC. The condenser is used to condense water vapor from the fractionator which is?? collected in the accumulator, and routed to the recycle water head pot. Then any water vapor is discharged through the stack.	iodine remaining in the feed in the acid fractionator overhead vaporizer before discharge to the atmosphere through the stack. The addition of sugar for the vaporizer also suppresses ruthenium volatility if any. The control of minimum acidity in the reagent streams is to prevent plutonium polymerization using the redundancy in the acid strength control devices.
Recovered acid storage tank	It stores recovered acid from the bottom products of the acid fractionator.	It measures about 7'4" diameter × 18'2" high [NUREG-0278].
Solvent batch stripping tank	It collects all organic wastes and acts as a central point. Types of chemicals used to strip the fissile materials and to decontaminate the organic waste vary. Repeated separate contacting with nitric acid and sodium carbonate is sufficient to decontaminate the solvent; however, oxalic acid and caustic can generally be used if additional and more radical treatment is required. The aqueous waste solutions from the solvent treatment are transferred to the GP concentrator feed tank in HILC.	It measures 3'9" diameter ×7'10" [NUREG-0278].
Solvent burner feed tank	It receives treated waste solvent from the stripping tank. It has a capacity of ~4300 gallons. Once it is full, it feeds to the solvent burner.	It measures 8'2" diameter × 16'6" high [NUREG-0278].
Solvent burner and quench pot	The burner is operated at 1550 ˚F, using propane gas. Once the combustion chamber is maintained at this temperature for 30 minutes, the waste solvent flows into the burner. Any P_2O_5 produced in the burner is converted to phosphoric acid in the quench pot, and sent as ILLW.	Both solvent burner quench pot and waste solvent burner have a combined dimension of 5'11" ×19'10" high. Both temperature and pressures in the burner, its blower and quench pot and air flow are included in the monitoring parameters in the safety interlock system. If any of these parameters are out of the tolerance condition, the burner will shut down. The alarm system is used to provide alerts to the operators.

A.1.6 Uranium Product Cell (UPC)

This cell provides a purification of the uranium product stream. Basically the aqueous strip solution containing the uranium is concentrated from 0.3-Molar to 1.5-Molar uranium and adjusted with nitric acid to ~2.05 Molar, and is again preferentially extracted by another 30% TBP organic solution in a pulsed column. Before leaving the column, the organic solution is scrubbed with 0.01 Molar and 2.3 Molar nitric acid solution to remove extracted ruthenium and zirconium-niobium. Hydroxylamine nitrate and hydrazine are also added to the scrub solution to remove any residual plutonium. Uranium is stripped from the organic solution in another pulsed column, using acidified water (0.01 Molar HNO_3). The solution is concentrated by evaporation from 0.4 Molar to 1.5 Molar uranium. Finally, the concentrated uranium solution is passed

through silica gel beds to remove residual traces of zirconium-niobium. Uranyl nitrate product is analyzed and transferred to the UF_6 facility for shipment or conversion. This cell has a dimension of 31ft × 27 ft × 52 ft.   The major equipment for this cell is provided in Table A-5.

Table A-5 Major equipment* descriptions in UPC [AGNS 1975]

Equipment	Operations	Descriptions
Decanter	This removes organic from the feed to the concentrator and thus optimizes operability of the concentrator.	Two decanters (1CU and 2EU) exist in this cell. Each measures at 1'2" diameter × 4'6" high.
Concentrator with condenser and reboiler	This system provides an effective control of the uranium concentration in the feed, and maximizes the decontamination factor in the extraction cycle. Each concentrator contains three stripping bubble caps, trays and mist eliminator York mesh-421 or equivalent, vortex breaker in lower section.	The two reboilers are vertical thermosyphon vessels that heat the feed to the concentrators to 116 F for 1CU system and to 126 F for the 2EU system. The 1CU concentrator is about 17' overall, top section contains a 5'6" diameter × 5'3" high, and bottom section contains a 2'6" diameter × 16'6" high. There are two systems (1CU and 2EU). Both condensers are to cool 1CU concentrator overheads at 5 MTU/day to temperature of 110 F. Cooling water at 85 F is used. 1CU condenser measures at 19" diameter × 168" tube length. Note that concentration control is used to ensure no criticality event. The parameters monitored include solution density, feed flow, process liquid temperature, and steam pressure. The monitoring is done in the redundant safety instrumentation and control. The system is shutdown when 87% of critical concentration is reached. In addition, if a high-pressure or high-temperature condition is detected in the concentrator, the reboiler stream is cutoff automatically.
U product cooler	This cooler provides cooling to the end product prior to transfer to the catch tanks.	It measures at 1' × 7'6" high.
U product catch tank	This tank is used to collet uranium product.	It has a volume capacity of 1210 gallons, which measures at 5'6" diameter ×8' long.
U product sample tank	It holds collected U product solution for sampling.	It has a volume capacity of 1210 gallons, which measures at 5'6" diameter × 8' long. Eight-hour production run batches may be collected and sampled.
Silica Gel beds	These beds are the final treatment for separating Zr and Nb from the U solution	Each bed measures about 1'8" × 13' high. These beds are provided and are used alternately to permit continuous operation. If the processing rate of 5 MTU/day is used, the spent silica gel adsorbent unit is regenerated about once a week by flushing the bed with a 0.4 Molar oxalic acid. The regeneration solution is directed to GP concentrator in the HILC for oxalic acid destruction and concentration of the solution.

*All equipment is constructed using the 304 stainless steel.

A.1.7   Plutonium Product Cell (PPC)

This cell provides the purification of the plutonium process stream. Plutonium aqueous stream leaving the partitioning column (electro-pulse column in HILC) is re-oxidized to the extractable tetravalent state with nitrate and is preferentially extracted into the TBP organic solution in the pulsed extraction column off the second plutonium cycle. This resulting organic solution is further scrubbed with 1.0 Molar nitric acid solution to remove extracted ruthenium and zirconium-niobium in the top portion of the column. Then this solution passes through a strip column where tetravalent Pu is transferred to an aqueous stream of dilute (0.3 Molar) nitric acid. A 3rd plutonium cycle is repeated for extraction and scrubbed as in the second cycle to remove

any fission product. A 3rd strip column is used to achieve higher Pu concentration using hydroxylamine nitrate to convert Pu to the strippable trivalent state. A TBP organic scrub solution also is used to remove any uranium from the plutonium aqueous stream as it leaves the 3rd strip column. After TBP is removed, the final plutonium concentration from 60 g/liter to 350 g/liter is achieved in a concentrator. After analysis, the final plutonium solution is stored in the product slab tanks before sending to the PNSL. Because of the concentration level (much above the ANS-8.1 subcritical limit), tanks must be criticality safe. This cell has a dimension of 20ft × 27ft × 51.5ft. Table A-6 lists the major equipment in this cell.

Table A-6 Major equipment used in PPC [AGNS 1975]

Equipment	Operations	Descriptions
2A/2B columns	These columns provide 2nd extraction cycle. It receives feed from the 1BP surge tank in HILC. The resulting Pu concentration is 10-20 g/liter.	2A measures 3'5" diameter × 43'7" high, and 2B measures 3'5" diameter × 30'3" high. Note that the concentration of Pu in the organic solvent exiting the 2B column exceeds 0.01 g/liter, an alarm is initiated to alert the operator of upset conditions.
3A/3B columns	These columns provide 3rd extraction cycle. They receive feed from the 2nd extraction cycle Pu stream. The resulting Pu concentration is about 40-50 g/liter.	3A measures 3'5" diameter × 43'7" high, and 3B measures 4' diameter ×46'3" high. Same alarm is issued for the Pu concentration in the organic solvent exceeding 0.01 g/liter.
3P concentrator, reboiler	This concentrator provides Pu concentration up to 350 g/liter.	The combined dimension is 4'6"×13'9" high.
Pu product storage tanks	These tanks provide an interim storage before transferring the Pu product solution to the PNSL.	Each measures 2.25"×7'6"×11'4".

A.2   Fuel Receiving and Storage Station (FRSS)

This station is designed to receive, handle and temporarily store spent fuel assemblies from nuclear light-water power reactors (see Figure A-2 to Figure A-7). A detailed top view and side view of the FRSS are shown in Figure A-13 and Figure A-14, respectively. Spent fuel assemblies are received in shielded casks by either truck or rail and delivered to FRSS in one of two bays as shown in this figure.

Figure A-14   Detailed top view of the FRSS [NUREG-0278]

A-25

Figure A-15   Side view of FRSS [NUREG-0278]

The fuel storage pool consists of 2 cask unloading pools where casks are opened, 1 fuel storage pool, 1 failed fuel pool where leaking fuel assemblies can be canned or uncanned, and 1 fuel transfer pool where fuel assembly is transferred to RPC. A 135-ton capacity cask handling crane is used for moving casks in and out of the cask unloading pool. Analysis of the dropped cask accident in the Unloading Bay the Test and Decontamination Pit or Decontamination Pit indicated that the integrity of the pool recirculation system can be breached. The water could be drained by gravity flow from the rupture lines; however, the placement of these lines prevents the pools from draining below10 feet of water shielding over the storage fuel. An aluminum storage canister is used to accommodate 8 pressurized water reactor (PWR) or 18 boiling water reactor (BWR) fuel assemblies. This canister is designed to provide geometric spacing for criticality and to withstand highest handling height drop to another canister at the highest rated speed of the canister handling crane without canister distortion. Spacing between loaded canister compartments is limited to the multiplication factor ≤ 0.95.   In addition, administrative procedures limit the movement of more than one fuel assembly in either cask unloading pool or transfer pool at a time.

Underwater hoods are provided as a means of containing the gaseous fission products during storage in the fuel storage pool. Any gases collected by the hood will be transported to the off-gas treatment system. Fuel assemblies are unloaded and stored under water to provide cooling and shielding. The 12' water shielding is provided in the pools which limit the surface dose to a calculated 0.08 mrem per hour. Handling systems are designed with special limit switches and mechanical stops to prevent fuel from raising fuel higher than the design depth of the shielding water. Storage capacity is 360 metric ton uranium of fuel.

The pool water is channeled and treated to promote maximum clarity, to control temperature, and to minimize corrosion and radioactivity by using continuous filtration through 95% efficient 5 μm pore size filter elements, cooling in heat exchanges to hold water temperature below 105 ˚F and demineralization. All expected wet surfaces in the FRSS is covered by a stainless steel liner up to the expected highest water mark.

Note that the FRSS structure is safety related up to 25 feet above the berm level and non-safety related above this level. The conditioned ventilation air to the FRSS is provided from an independent unit and a major portion of the air is channeled to the analytical filter station, is gathered and released to the stack. Dropped fuel element accident has been included in the safety analysis report, which shows the immediate release is negligible. A list of the major equipment in the FRSS is provided in Table A-7.

Table A-7 Major equipment description in FRSS [AGNS 1975]

Equipment	Operations	Descriptions
Cask handling/unloading cranes	These cranes are used for cask operations including unloading cask contents.	It includes the supports and restrainers to be used in cask operations. One crane has a capacity of 135 tons.
Fuel storage canister crane	This crane is used in the canister operations.	It includes the supports and restrainers to be used in canister operations (15-ton capacity)
Fuel storage canisters	They are used to store fuel assemblies inside the spent fuel pool.	Each canister can accommodate 8 PWR or 18 BWR fuel assemblies. The design of the canister and the spacing in the pool accounts for criticality concerns.
Fuel transfer conveyer	It is used to transfer fuel assembly from FRSS to RPC for processing	It is a cart-track arrangement which transports fuel assemblies from a vertical position in the fuel transfer pool to a horizontal loading position in the RPC.
Spent fuel pools	These pools provide radiological shielding and cooling for the stored spent fuel. The failed pool is intended to handle the leaking fuel pins.	Spent fuel poolis divided into five pools consisting of two cask unloading pools (CUP), one fuel storage pool, one failed pool, and one fuel transfer pool. Normally, 12-ft of water shielding is provided for the pools.

A.3 Control Room Area/ Plutonium Nitrate Storage and Load-out

Both Control Room Area (CRA) and Plutonium Nitrate Storage and Load-out (PNSL) are located outside high radiation process area, where thick concrete structures are used to provide adequate shielding for radiation (see Figure A-2 to Figure A-7). The description of CRA is discussed first, followed by the description of the PNSL.

A.3.1 Control Room Area (CRA)

CRA is an important area that houses the process control instrumentation, display and monitoring panels for all safety-related instruments within the process facility. This area also serves as the communication center for which operators can be directed to perform manual functions. An electrical cable distribution room is below this area providing space for wire and cable routing (see Figure A-7).

A.3.2 Plutonium Nitrate Storage and Load-out (PNSL)

This operation is to receive plutonium nitrate (250g/liter Pu at 3-Molar HNO_3) from the interim storage tanks in the PPC and store in geometrically favorable storage tanks within two storage cells. A total of 24 such slab tanks (each has a capacity of 232 gallons) distributed into four modules in a cell. All safety related storage tanks are constructed with titanium to minimize corrosion and contamination. The stored nitrate solution is cooled by a filtered, chilled recirculating air system to maintain temperature at about 140 ˚F. The nitrate solution is loaded from the storage tanks in the Plutonium Nitrate Cell (PNC) into a 50-liter sample tank, to the 10-liter load-out tank, and finally to 10-liter capacity shipping containers. Optionally, the nitrate solution from PPC can be transferred directly to the 50-liter sample tank. The shipping

containers are decontaminated and stored (up to five containers) in the Plutonium Nitrate Storage Area (PNSA). Loading onto the truck for shipment is also done in this facility. Similarly, neptunium nitrate solution can be stored and processed in this facility.

A.4   Waste Tank Equipment Gallery (WTEG)

This gallery serves as the radioactive liquid storage processing facility, which houses processing equipment and utilities to store radioactive liquid waste. This building has an area about 30 m^2, divided into three main sections: (a) a cold area which is non-radioactive, (b) a hot area which is radioactive, and (c) an intermediate area [NUREG-0278].

The cold area houses the waste tank operation instrumentation, which is used to monitor liquid level, specific gravity, temperature and pressure of the tank content.

A.5   Waste Tank Cells (WTCs)

This cell contains two high-level liquid waste (HLLW) storage tanks and two intermediate-level liquid waste (ILLW) tank. Only one tank is used at the time. The other tank is a spare. These tanks form the waste tank cells. Radioactive liquid is transferred from the main process building to WTEG through underground pipe vaults and distributed to the tanks through underground lines via the diverter cell in the WTEG.

A.5.1   High-Level Liquid Waste

Each stainless HLLW tank is 16.5 m in diameter by 6.1 m high with a capacity of 300,000 gallons. Each tank contains 48 5-cm diameter cooling coils, 18 air-operated ballast tanks round the perimeter of the tank, 9 air-operated ballast tanks in the main part of the tank, 22 air lift circulators, 5 steam-operated ejector pumps, water-seal type pressure/vacuum relief system, external temperature sensors, and 10 instrument dip tubes to measure liquid level and specific gravity. This tank is contained within an underground, cylindrical, stainless steel lined (up to 5.5 m on the vault side), concrete vault, which measures 18.3 m diameter and 7.6 m high. The thickness of concrete for the vault is 1.2 m, 0.9 m and 1.7 m for the floor, side and top, respectively. The tank is sloped inside the vault to facilitate drainage in the unlikely event of a leak. A sump around the perimeter of the vault is provided for containing any leak. Figure A-16 provides a cut-away view of the HLLW tank section.
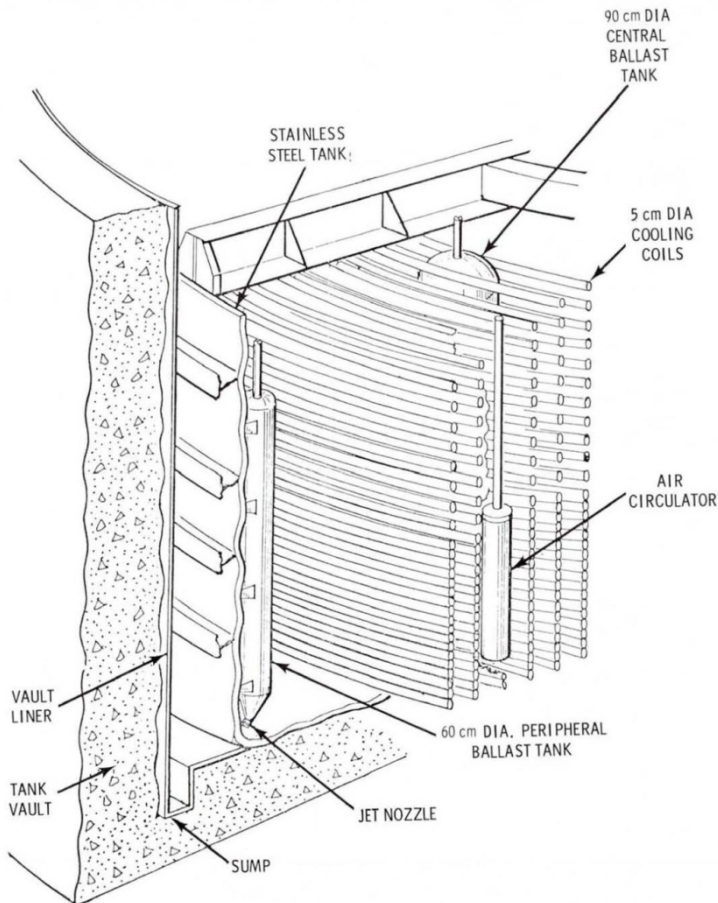
Figure A-16   Cut-away view of the HLLW tank section [NUREG-0278]

The stored HLLW will be maintained below 140 ˚F and will not be allowed to reach boiling temperature under any credible set of circumstances. The cooling in the tank is provided by the 48 cooling coils. Settling of the solids in the tank is prevented by using the ballast tanks, and air lift circulators. Any airflow through the waste solution tends to concentrate the solution by evaporation, which can amount to as much as 500 gallons/day/tank. If over-pressurization of the dissolve-off gas/vessel-off gas system occurs, which is an unlikely event, an automatic diversion of the HLLW off-gas to the UPC occurs, which will provide for de-entrainment of aerosols. Once the pressure surge has been relieved, normal venting of the HLLW-off-gas would be re-established.

A.5.2   Intermediate-Level Liquid Waste

The construction of the ILLW tank is very similar to the construction of the HLLW tank, except that ILLW tank does not have cooling coils, air lift circulators or ballast tanks, because the expected dose in the ILLW tank is much lower than that of HLLW. To minimize any solid settling, an air sparge system is used to agitate the waste. The feed for ILLW include the solvent treatment, laboratory, cell/equipment decontamination, blowdown from the acid fractionator overhead vaporizer and from solvent burner quench pot, off-gas scrubber solutions, HLLW off-gas condense, and silica gel regeneration from UPC. All of the ILLW solution is transferred to the tank from the GPW check tank in the main process building. All

interconnecting lines between the process building and the diverter, and between the diverter and the ILLW vault are enclosed within stainless steel lined concrete pip vaults. As in the HLLW, ILLW is equipped with instrument dip tubes and thermocouples.

The tank is designed to provide a narrow operating range with respect to the vapor phase pressure, which the vapor space operating pressure is not expected to be positive to the surroundings. Thus pressure operation is never allowed. The designed operating pressure is minus 3 inches of water to plus 10 inches of water.

A.6   Galleries

The galleries provide enclosure and protection for piping, process support equipment, and instrumentation components. These galleries are used by operating personnel and are used for many operations [NUREG-0278].

- Head Tank Galley/Pulser Equipment Gallery – They are used to house air pulsers for the solvent extraction columns, recovered acid tanks, recycled water tanks, and the cold off-gas blower. The galleries are located above, and provide access to, the process cells (see Figure A-5). They are constructed of reinforce concrete with 0.5 m thick walls.

- Piping and Instrument Gallery – This gallery contains piping, valves and process control instrument to serve the adjacent cells. It is located the north side of the process cells (see Figure A-3 and Figure A-4). As shown in Figure A-3, the lower piping and instrument gallery is identified.

- Filter Piping and Instrument Gallery (FPIG) – This gallery contains piping, valves and process control equipment necessary for the operation of the adjacent cells. It also contains contact cell ventilation air filter housing, Sample and Analytical Cell filter housings, Plutonium Product Cell filter housing, the 2.7 atmosphere steam system, and miscellaneous pumps (see Figure A-2)

- Crane and Equipment Maintenance (CEMG) Gallery – This gallery serves the crane maintenance for the RPC. A removable shielding door divides between this gallery and RPC (see Figure A-4). This is also used for equipment items entering and leaving RPC. This gallery is also interfaced with RMSC below through floor shielding plugs that occupy roughly about half of its floor space. A shielding personnel access door, a workbench, lifting device for the floor plugs, crane maintenance access platform, and an ultrasonic cleaning tank (0.5 m3 capacity) can be found in this gallery.

- Service Concentrator Gallery – This gallery houses service concentrator equipment, which is isolated by concrete walls and an airlock and is located on the southwest corner of the lower viewing and operating station (see Figure A-2). Both floor and walls are covered with acid-resistant paint up to a height of 0.9 m. Floor drains are provide to the feed tank to route any leakage back to the service concentrator or for transfer to the general purpose concentrator.

- Hull Monitor Maintenance Gallery – This gallery is used to maintain hull monitor and drive assembly, and is located between the PRC and RMSC, and is accessed from the lower viewing and operating station (see Figure A-2).

A.7   Stations

Stations are locations around the process cells that provide various functions for the facility. The major stations are described below [NUREG-0278]:

- Cask Loading Station (CLS) – This station is located east of the RMSC (see Figure A-4, Figure A-6 and Figure A-7), which is used to remove loaded solid scrap disposal containers and to secure them in the shipping cask for transport to the onsite underground solid waste storage area. Empty disposal containers also enter the RMSC from this station. Decontamination of the shipping cask is also done in this station.

- Viewing and Operating Stations – These stations are located on the south side of the RPC and RMSC (see Figure A-2 to Figure A-5). These stations are:

  o   Lower (LVOS) – It has an area of ~4.9 m wide which bounds the RMSC on the south side and west end, and the lower portion of the RPC on the south. Its function is to load leached hulls in interim burial containers and remote maintenance of equipment. It also contains 4 view windows that are 0.6 m by 0.7 m lead glass, oil-filled shielding windows installed in the concrete wall for observing activities within the RMSC. At each view window there are two master-slave manipulator wall tubes and one periscope wall tube. Additional periscope wall tube is provided for reviewing the hull dumping operation. A port for passing items into or out of RMSC is located in the east wall of this station.

  o   Grade (GVOS) – It has an area of ~4.9 m by 40 m, separated from the south side of the RPC and CEMG by a concrete wall (1.5 m thick). This station is used to conduct remote maintenance for the CEMG and operation of fuel shear mechanism and subsequent transport of leached hulls to the hull dumper. Four view shield windows are located in this station to observe operations in PRC and CEMG.

  o   Analytical (AVOS) – It has an area of ~5.1 m by 55 m, located north of the SAC. The operations in the SAC are conducted from this station through shielded view windows and manipulators. This station also contains glove boxes for taking samples from the UPC, PPC, and ILC.

  o   Top (TVOS) – It is a 4-m diameter circular area in the cold chemical station. It provides top viewing of the RPC operations. A shielding window is provided on the station's floor. Four penetrations to the floor for supplying auxiliary services to the RPC.

- Contact Equipment Removal Station (CERS) – This station provides access into the process cells below (see Figure A-5). Materials may be installed in and removed from these cells through shielding hatches in the cell ceilings. This station is served by a 55 metric ton crane.

- Equipment Maintenance Station (EMS) - This station performs final decontamination and repairs where items from the cells are repaired, prepared for transferring out of the building, or packaged for transfer to the underground waste storage. Occasion maintenance activities and repair of equipment can be done in this station. Concrete floor plugs provides access to the CEMG. This station contains a 55 metric ton crane for any major lifts as needed. This station is located above the CEMG and is located the top floor of the process building (see Figure A-5).

A.8 Sample and Analysis Cell (SAC)

This cell is used to provide a shielded area for remote sampling and analysis of highly radioactive materials from the process cells. It contains eight compartments, each having master-slave manipulators and viewing windows (see Figure A-5). Each compartment has a floor drain leading to the general concentrator for liquid waste, and a stainless steel liner to the floor and part of the walls. A power hoist is used to transport large items of equipment from compartment to compartment. The following functions are identified for the compartments:

- Two compartments contain samplers and equipment for remote sampling of the process streams and tanks.

- One compartment contains sample storage capability and equipment for disposal of surplus samples to a recovery tank.

- Two compartments contain equipment for remote radiochemical analyses.

- One compartment is used to provide decontaminated samples and to prepare weighed dilutions of samples to be analyzed in the HCLA.

- One compartment provides equipment for processing chemistry studies.

- One compartment has facilities to transfer equipment to and from the cell via a transfer drawer, and a scrap chute to the RMSC for disposal of canned solid waste.

A.9 Support Systems

This section describes the support systems used at the facility. These systems include the ventilation system, electrical power system, fire protection system, water reservoir, and laboratory area (hot and cold).

A.9.1 Ventilation System

This system provides pretreated airflow to the processing areas, maintains pressure differentials between zones as required to confine radioactivity to designated locations. The ventilation systems are designed to confine and channel any airborne release to treatment systems such as filters. Cooling is provided by the ventilation systems for the process cells. Figure A-17 shows the air flow diagram of the ventilation system. As shown in this figure, the air circulation for the FRSS is about 84,000 cubic feet per minute (CFM) and the process building air circulation is about 92,000 CFM. Table A-8 provides some operating conditions of the ventilation systems.
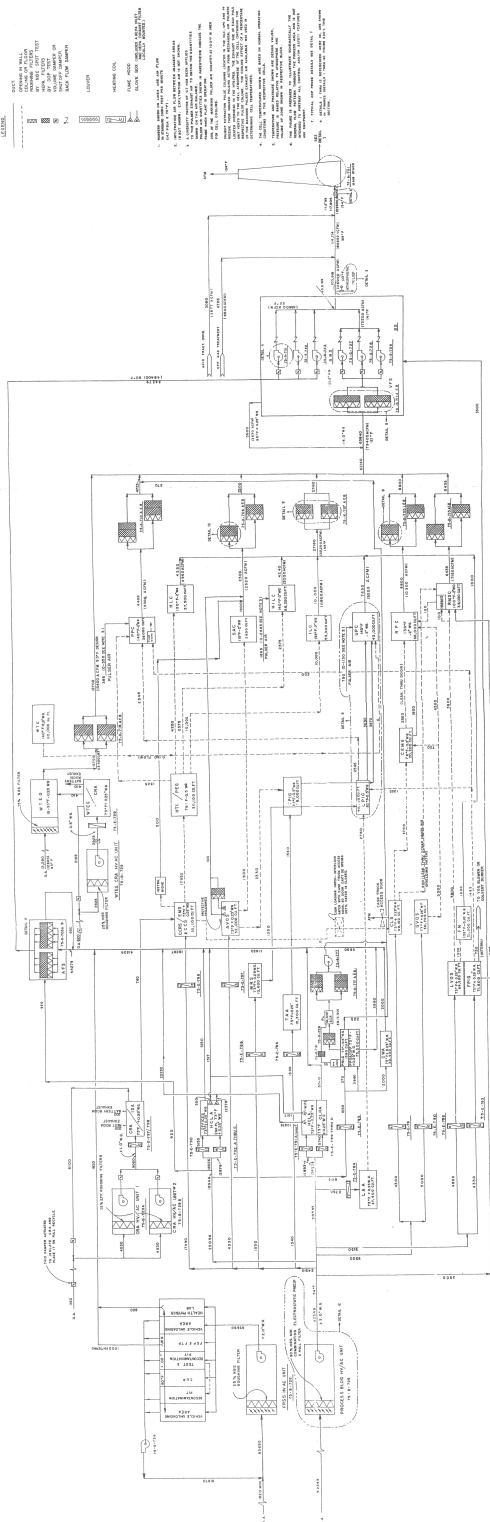
Figure A-17   Air flow diagram in the ventilation system [AGNS 1975]

Table A-8   Operating conditions of the ventilation systems at selected locations (see Figure A-17)*

Location	Temperature (˚F)	ΔPressure (inches of water gauge)	Zone Volume (ft³)
Remote Process Cell	130	-2	68,000
Remote Maintenance and Scrap Cell	100	-2	36,000
High Level Cell	130	-2	37,500
High Intermediate Level Cell	140	-2	56,200
Intermediate Level Cell	135	-2	53,300
Uranium Product Cell	143	-2	43,500
Plutonium Product Cell	140	-2	28,200
Waste Tank Equipment Gallery	93	-0.25	--
Waste Tank Cell	140	-2	110,000
Crane and Equipment Maintenance Gallery	76	-0.5	20,200
CERS/EMS	73	+0.25	33,100

*Both temperature and pressure shown are design values. ΔPressure is based relative to atmosphere and volume of zone shown is respective location.

According to [AGNS 1975], the supply system fan is sized to produce a total differential head of 6 inches of water gauge and deliver air at its discharge at +2.7 inches of water gauge. In the event the supply unit is off and the process building exhaust blowers continue to operate, the areas of the process building that are normally slightly positive will see negative pressure somewhere between atmospheric and -6 inches of water gauge. There are a total of three exhaust blowers. One is in stand-by mode. It would start up automatically on low negative pressure at the outlet of the primary HEPA filter bank in the process building. Figure A-18 shows the blower station building. As shown in this figure, this building contains the final filter banks, which are enclosed inside a concrete room. It also houses the process exhaust blowers, analytical exhaust blowers. The process building ventilation supply is also shown.
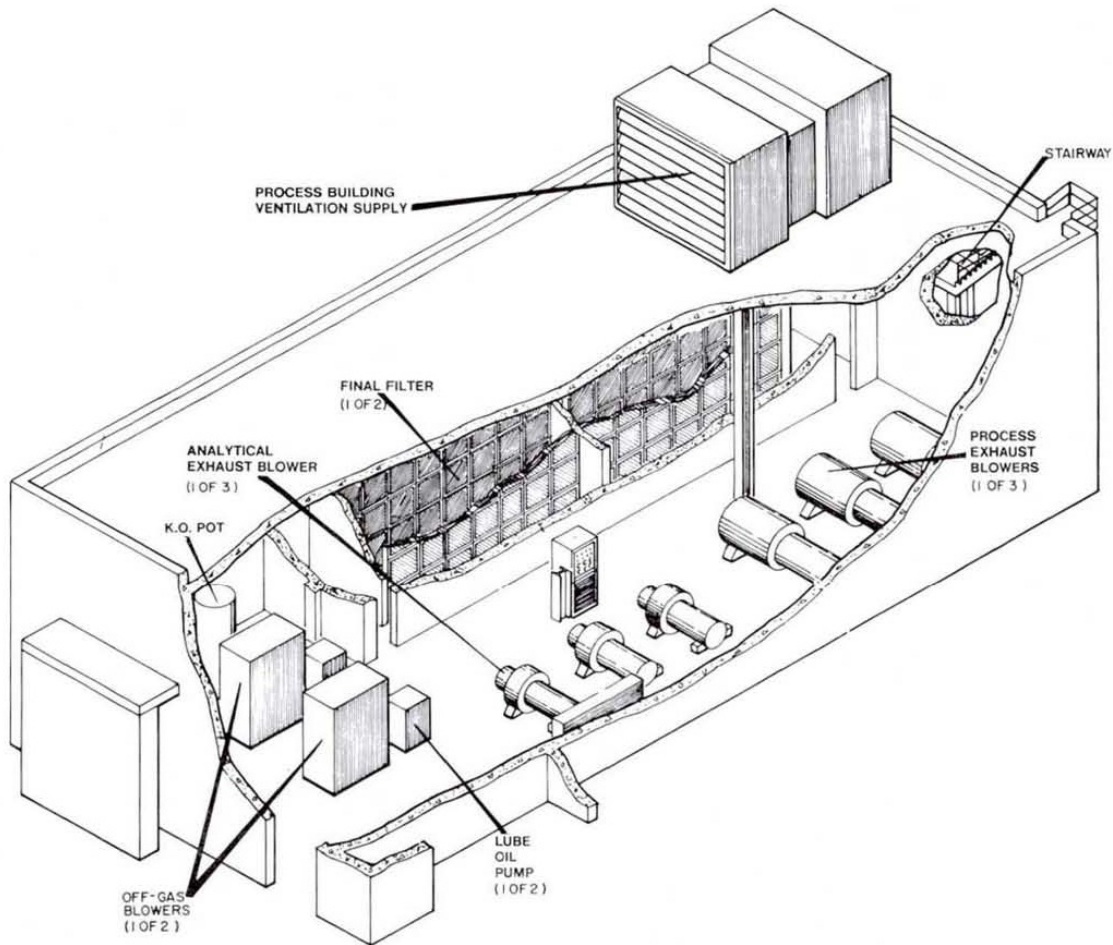
Figure A-18   Blower station building [NUREG-0278]

A.9.2   Off-Gas System

This system provides treatment of off-gases to remove contained particulates, iodine and oxides of nitrogen. There are two major off-gas systems: a dissolver off-gas (DOG) and vessel off-gas (VOG). The scrubbed and filtered gaseous effluents from these systems are released to the atmosphere via the stack (see Figure A-19). As shown in Figure A-19 [NUREG-0278], there are two types of off-gas systems (DOG and VOG).   Note that this figure is slightly different from what was reported in [NUREG-0278], because solidification processes were not included in the original BNFP. Each of the two systems is described below:
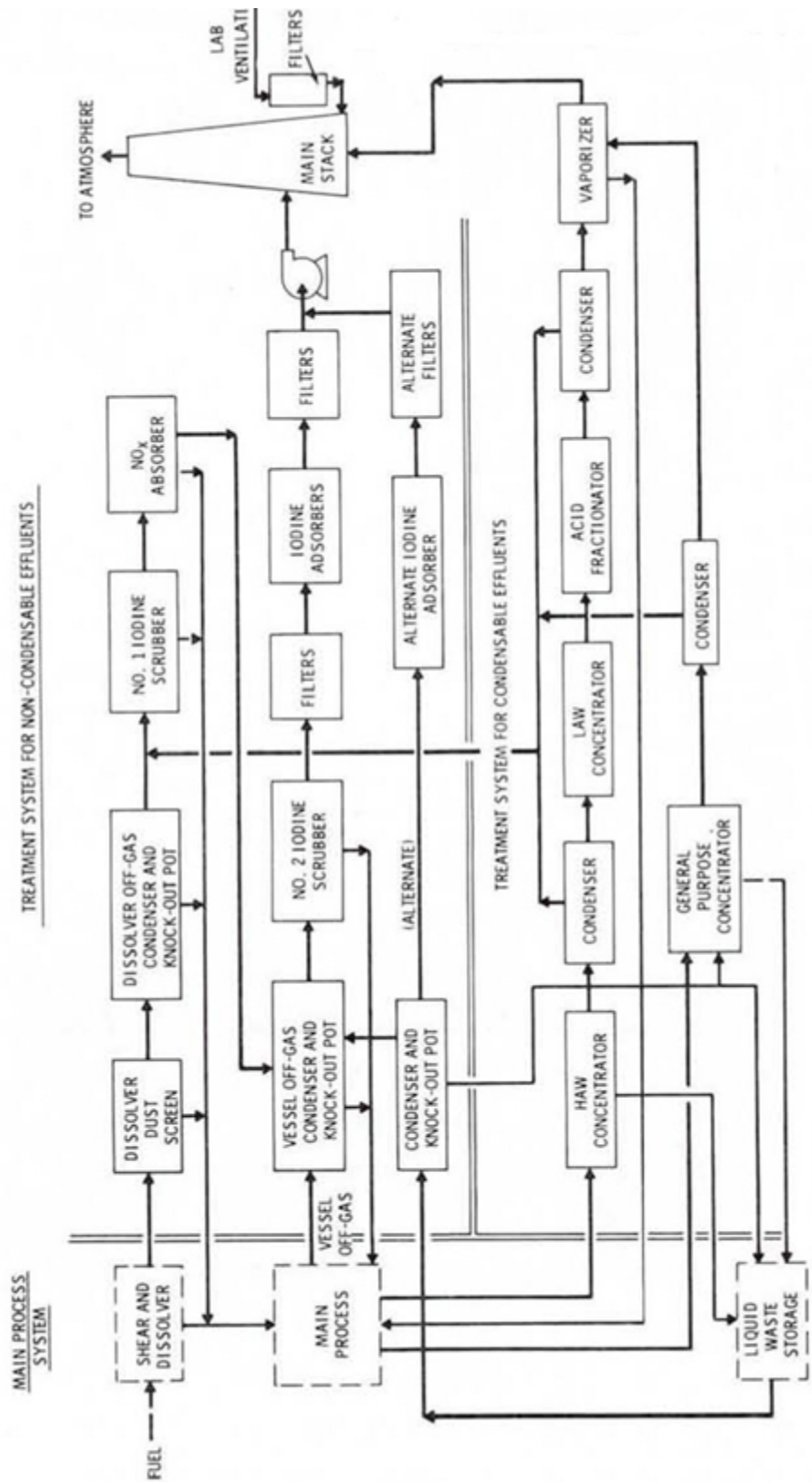
Figure A-19 Major off-gas systems

The DOG system provides treatment of off-gases from the dissolvers, high-activity waste concentrator condenser, plutonium vent knockout pot, acid fractionator condenser, and general purpose concentrator condenser. It contains an iodine scrubber, which uses the circulating mercuric nitrate-nitric acid solution to remove radioactive iodine. Since of the nitrogen oxides are primarily in the dissolvers, the NO_2 absorber is included in the DOG.

The VOG system provides treatment of off-gases from the high and intermediate-level liquid waste storage tanks, instrument purges, samplers, air lifts, sparges, steam from blowing or venting jets, and the waste solvent burner.

A.9.3 Electrical Power System

Normal electrical power is fed to the plant from the commercial substation. In the absence of the normal power, the emergency power is supplied by two independent diesel engine-driven generators. Within 10 seconds of normal power loss, both generators will start automatically. DC power is provided for instrumentation in the main control room and the waste tank equipment gallery area.

A.9.4 Fire Protection System

The source of the water for the fire protection system is based on the 60.5×10^6-gallon Beacon Pond in the area of the plant [AGNS 1975].

The process cells are protected by individual safety automatic fire detection and suppression system in each cell: Uranium Product Cell, Intermediate Level Cell, High Level Cell, High Intermediate Level Cell, and Plutonium Product Cell. Each cell is provided with a dual fire detection system consisting of one smoke detector and one combination rate-of-rise and fixed-temperature detector. The suppression system uses Halon 1301 as the fire suppressant agent.

Other areas in the plant are protected by the fixed wet-pipe sprinkler systems, which include the storage building, galleries, pump house, analytical filter station and ventilation filter station.

A.9.5 Hot and Cold Laboratory Area

This area contains 13 individual laboratories (two-story complex) that are equipped to provide specific types of analyses or services. One laboratory is equipped for receiving samples from the Sample and Analysis Cell. Both fume hoods and glove boxes are located in the laboratories. Waste drains from the sinks, fume hoods and glove boxes are routed to the laboratory waste catch tank or fume hood drain tank. A special α laboratory includes two special glove boxes for confinement of concentrated α-emitted radionuclides.

APPENDIX B

SELECTED PAST CHEMICAL EXPLOSION ACCIDENT CONDITIONS AND POSSIBLE EXPLOSION CALCULATION INPUT MODELS

This appendix provides insights gained from chemical explosion accidents that occurred in spent fuel reprocessing facilities in the past. Of particular importance is understanding of the chemical explosions in process vessels used in the operations of the reprocessing facilities.

Chemical explosions at a spent fuel reprocessing facility are most likely to occur in the operations involved with evaporation and concentration processes. The reason is that external heat is applied to remove unwanted liquid. The combination of heat and operation errors during transfer of TBP into the process vessel, where nitric acid and metal nitrates are present resulted in the "red oil" explosion condition. Lux and Vail [Lux 1995] classified the process vessels into 4 types: feed tank, sump receipt tank, mixer settler, and evaporator. Among these types, feed tank, sump receipt tank and mixer settler are categorized as unheated vessels. They are unlikely to yield a red oil explosion condition, because no heat is applied. Thus their accident frequency is $< 1 \times 10^{-6}$/yr. Unlike the unheated vessels, the evaporator, concentrator or denitrator vessel is subjected to external heat. If an operation errors results in unwanted TBP in the heated vessel, the red oil explosion condition is possible. Therefore, the accident frequency of this heated vessel is likely higher than that of the unheated vessel.

A selected set of the past explosion accidents are described in this appendix, along with details on the accident conditions, particularly for the solution states before explosions. An estimate of the total energy of the explosion is calculated. The description should be sufficient to allow the simulation of the accident through the use of explosion computer codes.

B.1   TNX Evaporator Nitration-Oxidation-Reaction Event (1953)

On January 12, 1953, a TNX evaporator exploded during the concentration of a uranyl nitrate-nitric acid solution. The TNX Semiworks program involved testing of equipment using the PUREX extraction operations. The Extraction solution included uranyl nitrate, nitric acid and 30%/70% TBP/Amsco [Colven 1953, Durant 1988]. Amsco is a kerosene-like diluent. As a part of the testing, all aqueous uranyl nitrate fed to the evaporator had at one time been in contract with this organic solvent. At the time of the explosion, the operator heard a rumbling noise and then observed orange-colored fumes evolving from the top of the column with a high-pitched roar. The explosion destroyed the evaporator by ripping it into 6 major pieces. Most of the damage was located in the lower half of the evaporator, including the pot. In the column, the bottom tray was bowed downward more than 1 inch, while other trays were bowed to a lesser extent - some upward and some downward. This explosion resulted in extensive damages to the building, including the sheet metal roofing and siding that were extensively dislodged throughout the building. It also resulted in two minor injuries of personnel.

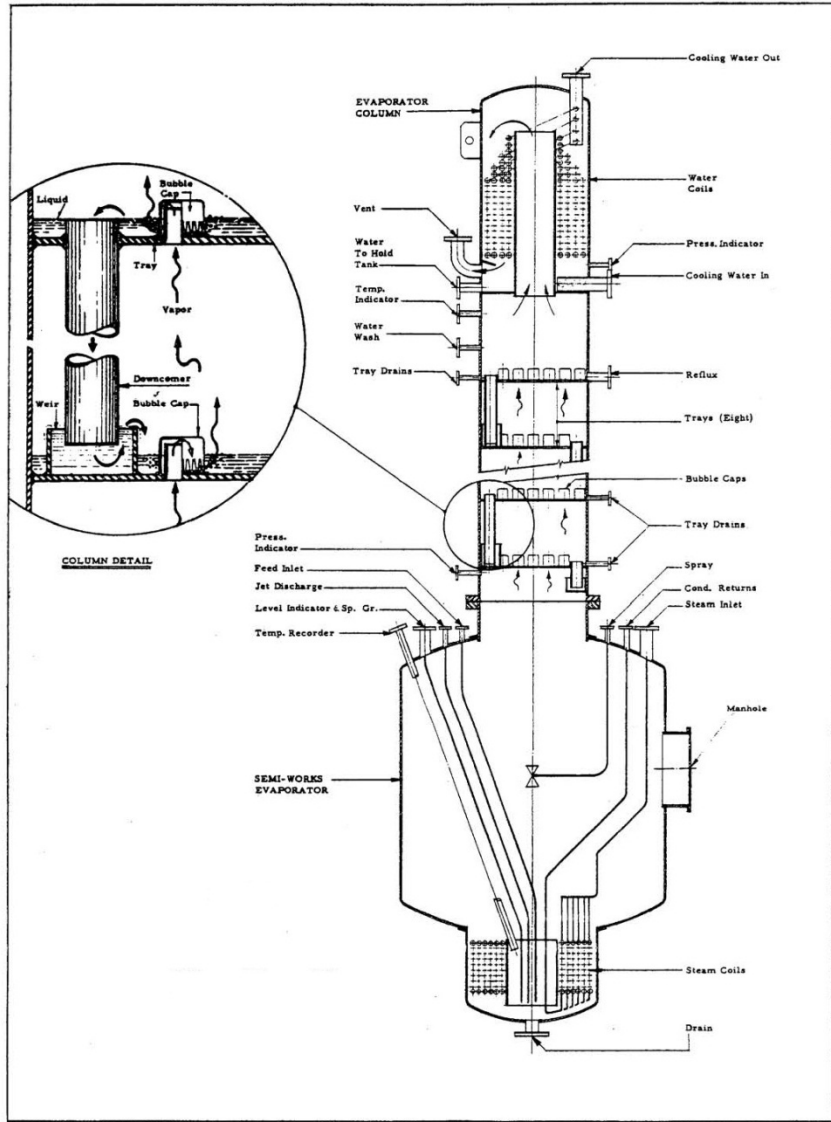The evaporator cross section is shown in Figure B-1, and its components and dimensions are shown in Table B-1.

Figure B-1   Cross section view of TNX evaporator [Colven 1953]

Table B-1 Description of the TNX evaporator [Colven 1953]

Parameter	Dimension
Evaporator Shell Diameter Height Dished Ends Wall Thickness Volume*	 6 feet (1.83 m) 5 feet (1.52 m) 1/4 –inch (0.64 cm) stainless steel 3/16 –inch (0.48 cm) stainless steel sheet (welded) 141.37 ft^3 (1057.52 gallons, 4 m^3)
Evaporator Pot Diameter Height Wall Thickness Heating Coils Volume*	 3 feet (0.91 m) 18 inches (45.72 cm) 3/16 –inch (0.48 cm) stainless steel 6 concentric banks of 1-inch schedule 40 stainless steel pipe 10.60 ft^3 (79.52 gallons, 0.3 m^3)
Column Diameter Height Number of Plates Humber Bubble Caps per Plate Wall Thickness Volume*	 2.5 feet (0.76 m) 16.5 feet (5.03 m) 8 30 3/16 –inch (0.48 cm) stainless steel 80.99 ft^3 (605.85 gallons, 2.29 m^3)
Total capacity Shell and Pot Column	1825 gallons (6.91 m^3) 1300 gallons (4.92 m^3) 525 gallons (1.99 m^3)

*Calculated based on the diameter and height of the component.

The cause of the accident could have been due to the fact that the liquid temperature recorder was broken and since the required degree of concentration was beyond the range of the specific gravity recorder, the evaporation was carried out for a specific length of time based on judgment. Also none of the organic solvent was thought to have been present in the feed, but after the explosion, it was discovered that Amsco diluent was present in the distillate catch tank. The constituents thought to be present at the time of the explosion are shown in Table B-2. This solution is based on the fourth and final charge consisting of the 70 gallon heel of the original solution and 160 gallons of previously evaporated material which had been diluted with water. After the explosion, it was noted that the column bubble-trays revealed partially plugging with solid materials. This partial plugging of the column could well have facilitated a pressure buildup in the evaporator prior to its rupture.

Table B-2 TNX explosion solution contents and conditions [Colven 1953]

Parameter	Values
Volume of Solution (liquid level recorder)	230 gallons (0.87 m^3)
Composition (analysis of feed)	UN-44 wt%, HNO3 -2.4 wt%
Specific Gravity (specific gravity recorder)	1.56*
TBP Content (from phosphorous analysis of residue after accident)	80 lbs (36.29 kg)
Amsco Content (found in distillate)	10-15 gallons (0.04 – 0.06 m^3)
Pot Maximum Temperature (calculated)	164 ˚C

*At the time of the explosion, the specific gravity recorder had gone off scale (> 2.0)

To model this explosion, one may want to ignore the volume in the column to simulate the plugging of the bubble-trays in the observation. Therefore, the model volume should be 1300 gallons, and the solution occupied about 230 gallons, which results in 1070 gallons for the gas headspace (see Table B-3).

Table B-3 Model input for TNX evaporator explosion

Parameter	values
Evaporator volume and dimension Volume (excluding Colum) Solution Gas headspace	 1300 gallons 230 gallons 1070 gallons
Solution (1) $UO_2(NO_3)_2$ + HNO_3 Volume Mass* $UO_2(NO_3)_2$ Mass** Molar HNO_3 Mass** Molar Solution Density* Viscosity$^\$$ Surface Tension$^\$$	 230 gallons (0.87 m^3) 1.26x10^6 g 5.97x10^5 g 1.74 3.26x10^4 g 0.6 1.56 g/cc 3.05 mPa-s 77 mN/m
Solution (2) TBP/Amsco TBP Mass Mole[1] Energy[2] Amsco Volume Solution Volume Density[3] Viscosity[3] Surface Tension[3]	 30 vol%/70 vol% 80 pounds (36.29 kg) 130.4 5.7581x10^7 Joules 10-15 gallons (medium: 12.5 gallons or 4.73x10^{-2} m^3) 17.9 gallons (6.78x10^{-2} m^3) 0.77 g/cc 1.2 mPa-s 22 mN/m

*Calculated using the given volume and the specific gravity of 1.56 provided in Table C.1-1.
**Calculated using the wt% given in Table C.1-1.
$^\$$Adapted from Table II and Table III of Ref.[Ondrejcin 1961] for 2 M UN solution.
[1]Molecular weight of TBP assumes to be 278.28 g/mole
[2]Assumed an energy of 4.4158x10^5 J/mole at 120 ˚C [Hyder 1994]
[3]These properties are approximated values from Appendix B of Ref. [Klophenstein 1958].

B.2   A-Line Denitration Explosion (1975)

The A-Line facility in F Area at the Savannah River Plant began operation in 1954 [McKibben 1976]. The facility's function is to convert uranium nitrate (UN) solution to UO_3 powder. The process flow diagram is shown in Figure B-2. As shown in this figure, the overall processes can be summarized by the following:

1.   Solvent decantation – to move organics (such as 30% TBP and 70% n-paraffin diluent) from the UN solution before it is sent to the evaporation operation.

2.   Evaporation – to concentrate the UN solution to increase the uranium concentration in the evaporators.

3.   Purification with silica gel – to reduce the ^{95}Zr and ^{95}Nb content in the UN solution from 2 above before sending to the hydrate process in the silica gel columns.

4.   Evaporation to hydrate – to concentrate UN and water to corresponding uranyl nitrate hexahydrate (UNH) or $UO_2(NO_3)_2 \cdot 6H_2O$ by evaporation in the hydrate evaporators before sending to the denitrators.

5.   Denitration – to converts UNH to UO_3 by thermal decomposition in the denitrators.

Figure B-2   A-Line facility process flow diagram [McKibben 1976]

The layouts of the equipment and facility are shown in Figure B-3 to Figure B-5. As shown in these figures, there are a total of 6 denitrators located on the first floor in the facility. The material inputs to these denitrators are provided by the 3 hydrate evaporators. Ultimately the silica gel columns provided the feed for the hydrate evaporators.

Figure B-3 Vertical sectional elevation view of the A-Line facility [McKibben 1976]

Figure B-4   First floor view of the A-Line facility [McKibben 1976]

Figure B-5   A-Line denitrator assembly [McKibben 1976]

The explosion and fire accident on February 12, 1975 in the facility was due to a number of mishaps in the operations prior to the accident [McKibben 1976]. When the facility was shut down a month before the accident, one of the silica gel columns was regenerated by flushing with 14,000 liters of dilute nitric acid before sending to the concentrate hold tanks (S-1-8 and S-1-9), which resulted a nominal uranium concentration of 250 g per liter and a specific gravity (SG) of 1.3, compared to the normal concentration of 400 g per liter and SG of 1.53. Normally, the TBP may have been present in an organic layer atop the silica gel column (at U concentration of 250 g per liter). With this flushing the resulting organic adduct became the $UO_2(NO_3)_2 \cdot (TBP)_2$ or TBP-UN. At the concentrate hold tanks, the aqueous solution (UN) is normally heavier than the organic TBP, when the UN is sent to the hydrate evaporator(s) then to the denitrator(s). However, because the resulting adduct at U concentration of 400 g per liter is heavier than the aqueous solution (UN) compared to a U concentration of 250 g per liter as shown in Figure B-6, the adduct was sent to the hydrate evaporators then to the denitrators. As shown in this figure, UN is heavier than adduct at the U concentration of 250 g per liter.
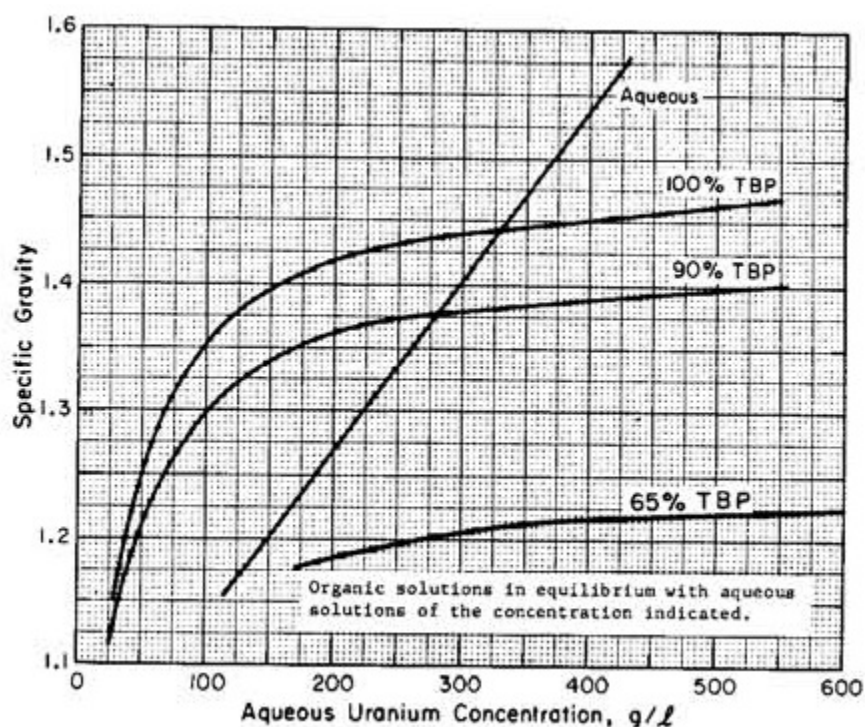

Figure B-6   Specific Gravity (SG) for the A-Line Solution at 25 ˚C [McKibben 1976]

On February 11, 1975, these adducts made their ways to the denitrators. Noticeable foaming was observed at 200-300 ˚C instead of at 500 ˚C [McKibben 1976]. On February 12, 1975, operators added a commercial silicone antifoam agent to denitrator pot C-3-4 and started its pot agitator. The temperature of the pot was increased to 400 C. Approximately 2 minutes later two operators in the denitrator room observed a puff of red-brown smoke from this pot. They immediately turned off the heat to C-3-4 and turned on an emergency off-gas system to provide extra exhaust capacity for the pot. Explosion and fire occurred afterward. Both of these operators received minor injury from the explosion. The destruction caused by the accident is shown in Figure B-7 to Figure B-11. As shown in these figures, there was considerable damage to the "Transite" walls in the building, but no damage was done to the building's structural supports. As shown in Figure B-7 to Figure B-10, the ejected materials from the exploded

denitrator pot have accumulated on the equipment, walls and pipes. Figure B-11 shows the external walls of the facility at the east side which have been blown out.
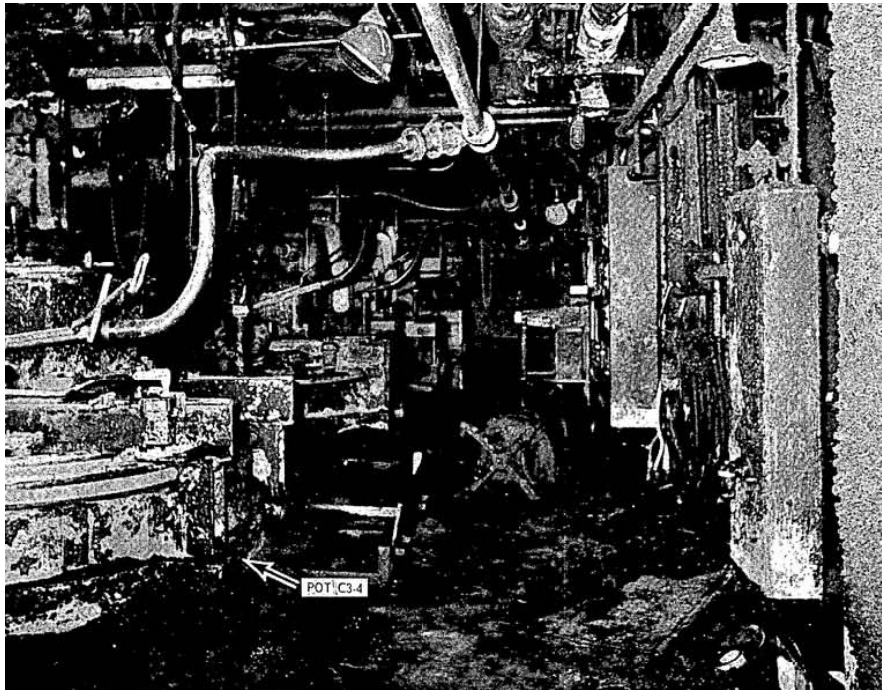

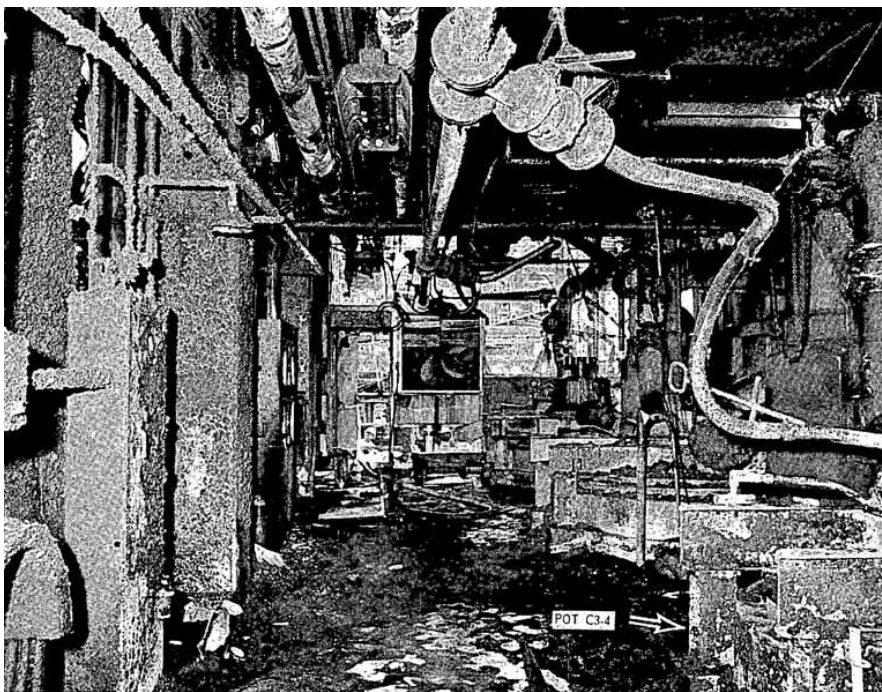**Figure B-7   North view of the damaged denitrator room after explosion [McKibben 1976]**

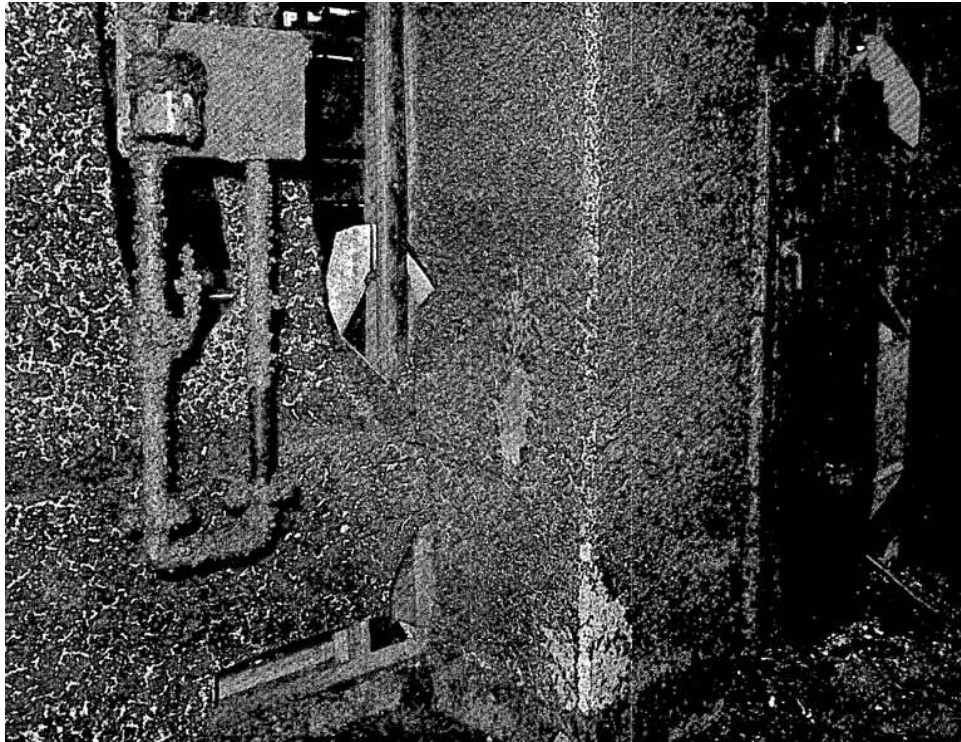**Figure B-8   South view of the damaged denitrator room after explosion [McKibben 1976]**

Figure B-9   Deposits on walls in denitrator room after explosion [McKibben 1976]

Figure B-10   Close-up of deposits on walls in denitrator room after explosion [McKibben 1976]

Figure B-11   View of the facility outside of east wall after explosion [McKibben 1976]

To construct the solution content at the time of the explosion, the following data is assumed and considered (see Table B-4):

1.   Approximately 14,000 liters of dilute HNO_3 added to S-1-8 and S-1-9 tanks, which yields 250 g U/liter at Specific Gravity (SG) of 1.3.

2.   Using Figure B-6 and 250 g U/liter, the following SGs for TBP and aqueous solution indicate that the TBP adduct becomes heavier than the aqueous solution, which is then sent to the hydrate evaporators and subsequently to the denitrators.

   a.   SG = 1.43 for 100% TBP
   b.   SG = 1.375 for 90% TBP
   c.   SG = 1.3 for aqueous solution

3.   Using Figure B-6 and 400 g U/liter, the following SGs for TBP and aqueous solution indicate that normally the aqueous solution is heavier than the TBP solution, so that only the aqueous solution is sent to the hydrate evaporators and subsequently to the denitrators.

   a.   SG = 1.45 for 100% TBP
   b.   SG = 1.38 for 90% TBP
   c.   SG = 1.535 for aqueous solution

4.   Approximately 120 liters of TBP-UN adduct were added to the C-3-4 denitrator pot prior to the explosion [McKibben 1976].

5.   Because of the temperature at 121°C, the TBP-UN adduct sent to the hydrate evaporators did not yield accident conditions.

6.   According to Figure B-5, the typical A-Line pot (denitrator) has a diameter of 6 ft. There were no additional dimensions given. This pot contains a motor drive for the agitator, which includes a plow. The bottom of the pot is round, but not hemispherical. Assuming one can scale the information from this figure, the deepest part of the pot is ~2.05 ft, and shadow part is ~1.42 ft. Then the average height is 1.74 ft. It is assumed that the volume of the pot can be thought to be a right cylinder with the given diameter and average height calculated: 49.2 ft^3 or 1.3932 m^3 (1.3932x10^3 liters).

7.   The total energy of the explosion is about 7x10^5 kcal or 2.93x10^9 J, and the shock pressure is ~30 to 40 atmospheres and temperature is ~4650 K [McKibben 1976].

8.   Using the data from 1,2 and 4 above, the mass of TBP is 1.43 x 120 liters = 171.60 kg, assuming 100% TBP. Using a molecular weight of 278.28 g/mole, it yields 616.65 moles.

9.   Using the specific reaction energy for TBP (4.4158x10^5 J/mole) given in Ref.[Hyder 1994] , the moles given in 8 above yields an energy of 2.72x10^8 J which is an order of magnitude smaller than that given in 7 above.

10. Using the upper bound specific energy range of (1610-1830 kcal/mole) for TBP as given in Ref. [Hyder 1994] and averaging this range, then the total energy is 4.44x10^9 J, which is slightly higher than the value given in 7 above.

11. Based on the calculations in 9 and 10 above, the possible specific energy for the TBP in this accident is given by 2.93x10^9 J/616.65 moles = 4.75x10^6 J/mole.

12. Based on the specification for each denitrator, a single batch contains about 2000 pounds of uranium or 907.18 kg [Harmon 1976].

13. According to the functionality of the hydrate evaporator, the goal is to produce UNH at 1.2 kg U per liter before sending to the denitrators [McKibben 1976]. Based on this density, and 12 above, the volume for the UNH in the denitrator should be 756 liters.

14. Total solution volume occupied by the TBP-UN and UNH is 756+120 = 876 liters.

15. Assuming the calculated available volume from 6 above occupied by machinery and piping of 10%, then the gas space in the denitrator is 1260-876 = 384 liters.

16. Since there was no specification of the material make-up of the denitrator given, it is assumed to be stainless steel with an average thickness of 0.25 inches.

17. Since there was no specification about the pot agitator, it is assumed to be stainless steel. See Figure B-5 for the location of the agitator and plow component.

18. The properties of UNH are assumed as [Lang 1968]:

   a. Density, according to 12 above is 1.2 g U/cc. Using the value of 1.2 g U/cc, the UNH density is 2.465 g/cc. However, at the accident condition, the aqueous solution is heavier than the organic, so it should be at 1.535 g/cc according to 3 above.

   b. Viscosity of 19 centipoises at 85 °C.

   c. Surface tension of 45 dynes per cm at 90-100 °C.

Table B-4 Input model for A-Line explosion

Parameter	Value
Tank Geometry and Information	
Diameter	6 ft (1.82 m)
Average Height	1.74 ft (0.53 m)
Thickness	0.25 inch (0.635 cm)
Volume	~1400 liters
Material	Stainless steel
Internal Component: pot agitator	Stainless steel
Solution (TBP-UN):	
Volume	120 liters
Mass	171.60 kg
Energy[1]	2.93×10^9 J
Density[2]	1.45 g/cc
Viscosity[2]	3.885 mPa-s
Surface Tension[2]	27.79 mN/m
TBP Concentration	80 mg/liter[4]
U Concentration	250g/liter[5]
HNO_3 Concentration	3 Molar
Solution (UNH):	
Volume	756 liters
Mass of U	907.18 kg
Density[3]	1.535 g/cc
Viscosity[3]	19 mPa-s
Surface Tension[3]	45 mN/m
HNO_3 Concentration	3 Molar[6]
Headspace Gas:	
Volume	384 liters

[1]See text on assumptions
[2]Assumed 100% TBP, and use values reported at 20 °C [Brennan 1951].
[3]See Assumption 18 above.
[4]Based on the solubility of the TBP in aqueous UN solution at 25 °C [McKibben 1976].
[5]Accident condition.
[6]Assumed to be same as TBP-UN.

B.3   TOMSK-7 Explosion (1993)

A Red oil explosion at the Siberian Chemical Combine in TOMSK-7, Russia on April 6, 1993 caused extensive damage to the nuclear fuel reprocessing facility and a large release of radioactive materials. This explosion was due to a runaway exothermic chemical reaction of a process vessel containing a concentrated solution of uranyl nitrate, plutonium nitrate, nitric acid, organics (Tributyl phosphate, TBP), and fission products [Rodriguez 1995]. It has been reported that there were two explosions, the first explosion was caused by the decomposition of

TBP in the nitric acid, and the second explosion was due to the flammable gases released from the first explosion, which caused extensive damages to the concrete structures which housed the exploded process vessel [Usachev 2003]. In order to model this accident, the information about the constituents in the process vessel, as well as the vessel dimensions and properties must be given.

The exploded process vessel is detailed in a Los Alamos National Laboratory (LANL) report [Rodriguez 1995]. The dimensions and its contents before the explosion, and mechanical properties of the vessel are given in Figure B-12. As indicated in this figure, the vessel is a cylinder with a cone-type bottom. To simplify the modeling of this accident, all inter-connection piping and internal can be ignored. Furthermore, the assumption of a right cylinder for modeling this vessel may be used. Table B-5 shows the content and thermal conditions of the vessel, and the calculated dimension for the vessel. The average thickness is calculated in this table. Since the vessel is made of stainless steel 304, and the exploded conditions calculated about the vessel breach [Rodriguez 1995], Table B-6 provides the properties to be used for modeling this vessel.
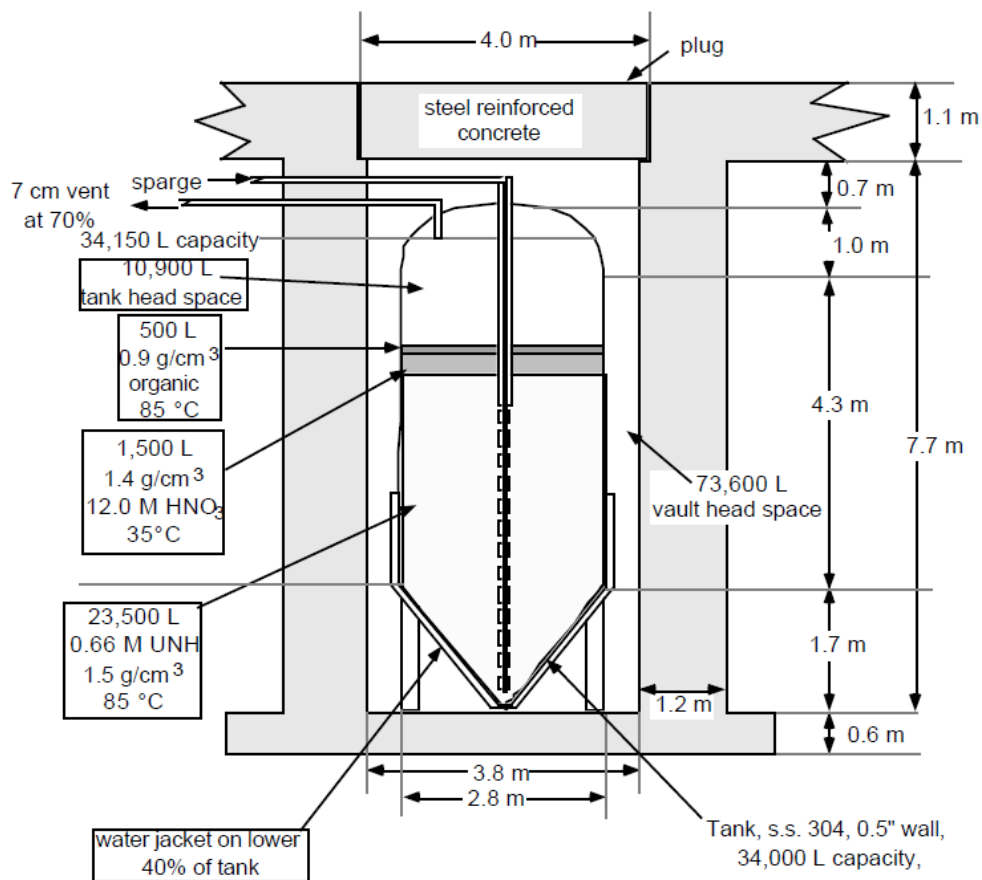

Figure B-12   Pressure vessel and surrounding structures, dimensions, and contents [Rodriguez 95]

Table B-5 Vessel contents and calculated dimensions from Figure B-12

Contents	Volume (liter)	Density (g/cc)	Temperature (C)
headspace	10,900	-	
Organic layer (TBP)	500	0.9	85
HNO3 (12 M)	1,500	1.4	35
UNH (0.66 M)	23,500	1.5	85
Calculated Vessel Dimension			
Available volume	36.4[A] m3	Inside radius	1.384[C] m
Average wall thickness	1.6[B] cm	Inside height	4.238[D] m

[A] The sum of the content volumes
[B] The average of the reported thickness between 14 to 18 mm [Rodriguez 1995]
[C] Calculated by subtracting the average wall thickness to the diameter of 2.8 m from Figure C.3-1.
[D] Calculated equivalent height, assuming the vessel is a right cylinder.

Table B-6 Stainless steel 304 properties [ASM 2012]

Property	Metric	English
Density	8 g/cc	0.289 lb/in2
Ultimate tensile strength	505 MPa	73.2 ksi*
Yield tensile strength	215 MPa	31.2 ksi**
Young's modulus (modulus of elasticity)	193-200 GPa	28000-29000 ksi***
Poisson's ratio	0.29	0.29

*In comparison to reported value of 70 ksi at ~40 ˚C, and 62 ksi at 135 ˚C for Gr.70 and 80 ksi at ~40 ˚C and 77 ksi at 135 ˚C of 304 SS [Rodriguez 1995]
**In comparison to reported value of 25 ksi at ~40 ˚C, and 20 ksi at 135 ˚C for Gr.70 and 35 ksi at ~40 ˚C and 26 ksi at 135 ˚C of 304 SS [Rodriguez 1995]
***In comparison to calculated value using Equation 12 in [Rodriguez 1995], the value is ranging from 27920-28170 ksi @ 100 ˚F, and 26670 ksi @316 ˚F.

Table B-5 lists the densities and volumes of the constituents in the vessel at the time of the explosion, which was based on the material flows into the vessel. The total content mass calculated from this table is 37,800 kg, and the bulk density is about 1.4824 kg/liter or 1.4824 g/cc. According to another LANL report [Eisenhawer 1995], there should be a total of five layers in the vessel before the explosion (see Figure B-13). As shown in Figure B-13, the reaction layer is not identified in Figure B-12, because information about this layer is not well known. However, brown NO_2 fumes were seen escaping from the plant stack [Eisenhawer 1995], which provided clues about this reaction layer. The following section discusses the possible reactions and release energy of the accident.
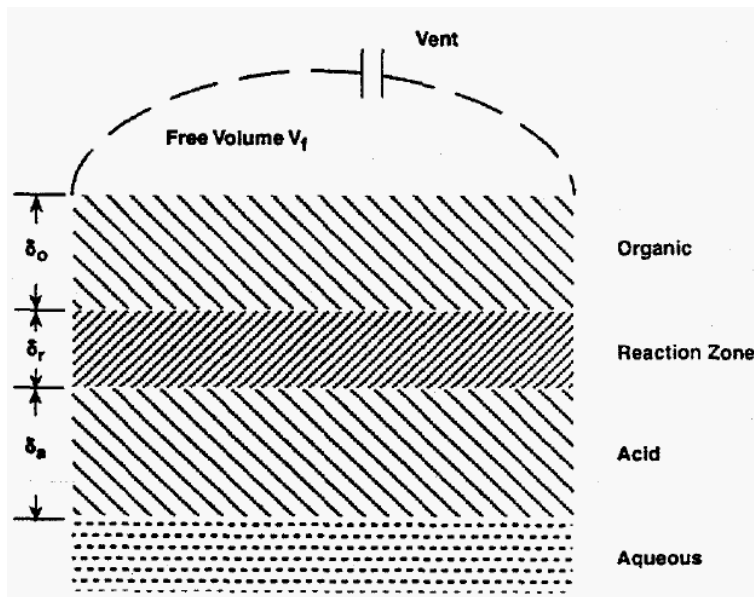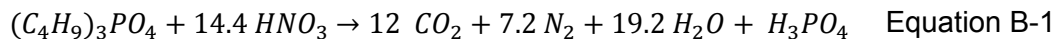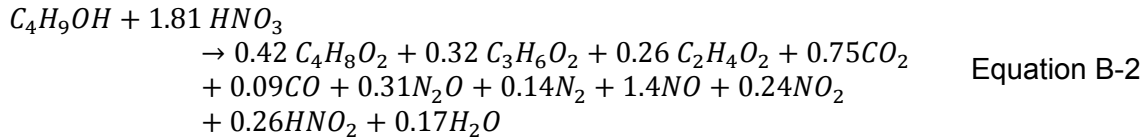
Figure B-13   Layers of contents in TOMSK-7 process vessel at accident time [Eisenhawer 1995]

At high temperatures, TBP tends to decompose, especially with nitric acid and other constituents. The reactions are complicated, and there are still many unknowns about the end products, which depends on the concentrations of the reactants involved, impurities, and temperatures. The complete oxidation of TBP by the nitric acid is illustrated below [Hyder 1994]:
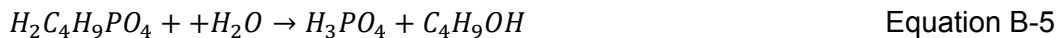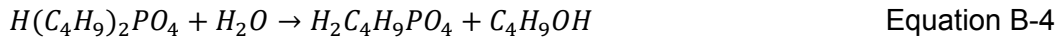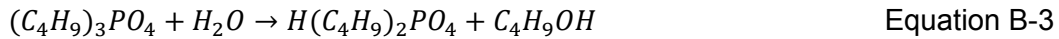
$$(C_4H_9)_3PO_4 + 14.4\ HNO_3 \rightarrow 12\ CO_2 + 7.2\ N_2 + 19.2\ H_2O +\ H_3PO_4 \quad \text{Equation B-1}$$

The total heat generation from this reaction is about 1610 – 1830 kcal/mole (6.7407×10^6 – 7.6618×10^6 J/mole) TBP, assuming the reaction takes place at 25 °C. The molecular weight of TBP is 278.28 g/mole. In the same report [Hyder 1994], the decomposition of TBP yields a heat generation rate of about 105.47 kcal/mole (4.4158×10^5 J/mole) at 120 °C based on a series of chemical reactions, considering the decomposition of TBP to n-butanol, and then the reaction of n-butanol with HNO3, etc. Because there was evidence of NO_2 release from the plant, the reaction in Equation (B-1) may not adequate to explain the NO_2 release.

Experiments on the reaction between TBP and nitric acid solutions have been performed by Smith [Smith 94]. In the experiments, Smith examined both single phase (organic) and two phase (organic and aqueous) TBP/HNO_3 reacting solutions at temperature above 100 °C. For closed systems, such as in TOMSK-7, the oxidation rate constant was about 1.33×10^{-3} per minute. In the experiments, Smith measured the product gases from the TBP/HNO_3 reactions, which include CO_2, CO, NO, N_2O, N_2 and NO_2. In a two phase experiment (#13) [Smith 1994], the following chemical reaction for the n-butanol (C_4H_9OH) and nitric acid, after the TBP saturated with 3.6 M HNO_3 and 10 M HNO_3 at 110 °C were mixed:

$$C_4H_9OH + 1.81\ HNO_3$$
$$\rightarrow 0.42\ C_4H_8O_2 + 0.32\ C_3H_6O_2 + 0.26\ C_2H_4O_2 + 0.75CO_2$$
$$+\ 0.09CO + 0.31N_2O + 0.14N_2 + 1.4NO + 0.24NO_2$$
$$+\ 0.26HNO_2 + 0.17H_2O$$

Equation B-2

The butanol can be formed by the hydrolysis of TBP in the following reactions [DNFSB T33]:

$$(C_4H_9)_3PO_4 + H_2O \rightarrow H(C_4H_9)_2PO_4 + C_4H_9OH$$

Equation B-3

$$H(C_4H_9)_2PO_4 + H_2O \rightarrow H_2C_4H_9PO_4 + C_4H_9OH$$

Equation B-4

$$H_2C_4H_9PO_4 + +H_2O \rightarrow H_3PO_4 + C_4H_9OH$$

Equation B-5

Using Eq. B-3, each mole of butanol would yield about 3 moles of gases. Based on the hydrolysis of TBP from Eq. B-4 to Eq. B-5, each mole of TBP would decompose to 1 mole of butanol. Knowing the number of moles of TBP, one could determine how much gases would be generated. Smith has correlated the gas generation with the organic to aqueous thickness ratio. Using Table B-5 for the organic and aqueous volumes measured for the TOMSK-7 accident, assuming the cross section area of both volumes are the same, then the organic to aqueous is 1/3. Reading from Figure B-14, the gas generation rate is about 0.01 liter per minute per M (M=moles of HNO_3/liter). As shown in Table B-5, the nitric acid layer is at 12 M. Therefore, the gas generation rate is about 0.12 liter per minute. It has been estimated that the total gas generated from the accident is about 250 m³ [Usachev 2003], which could be relatively high. Using the experimental results from Run#13 of Smith [Smith 1994], the initial organic density is ~1.065 g/cc, which utilizes about 3 moles of TBP per liter. Using this number of moles of TBP per liter, then the organic layer as described in Table B-5 would yield about 1500 moles of TBP. For each mole of TBP decomposed, there would be about 3 moles of gases generated. Therefore, about 4500 moles of gas generated. For an ideal gas, each mole of gas has a volume of 22.4 liters at the standard condition, which implies that the total volume generated by the organic layer is about 100 m³. In terms of the energy generation, the energy generated is about 1.0111x10¹⁰ J to 1.1493x10¹⁰ J, using the energy released from Eq. B-1, and the previously calculated number of moles of TBP. If using the energy generated suggested by [Hyder 1994], the total energy generated for the same number of TBP moles is reduced to about 6.6237x10⁸ J.

In terms of the fluid properties, [Bajoria 2012] published the TBP with nitric acid property plots (Figure B-15 to Figure B-18).

The first simulation for modeling this explosion should consider the input parameters as provided in Table B-7.
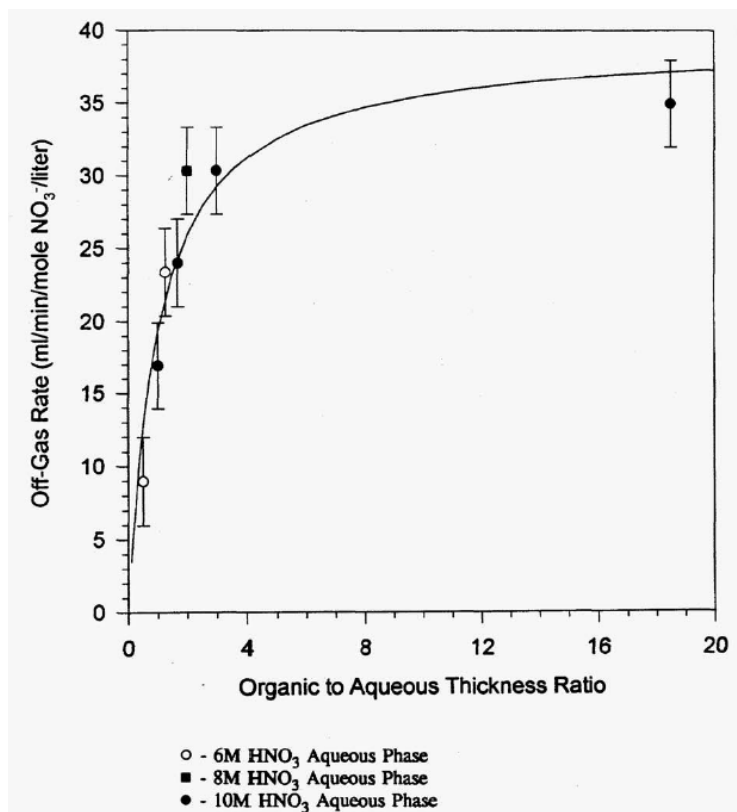
Figure B-14   Gas generation rate as a function of the organic to aqueous thickness ratio [Smith 1994]
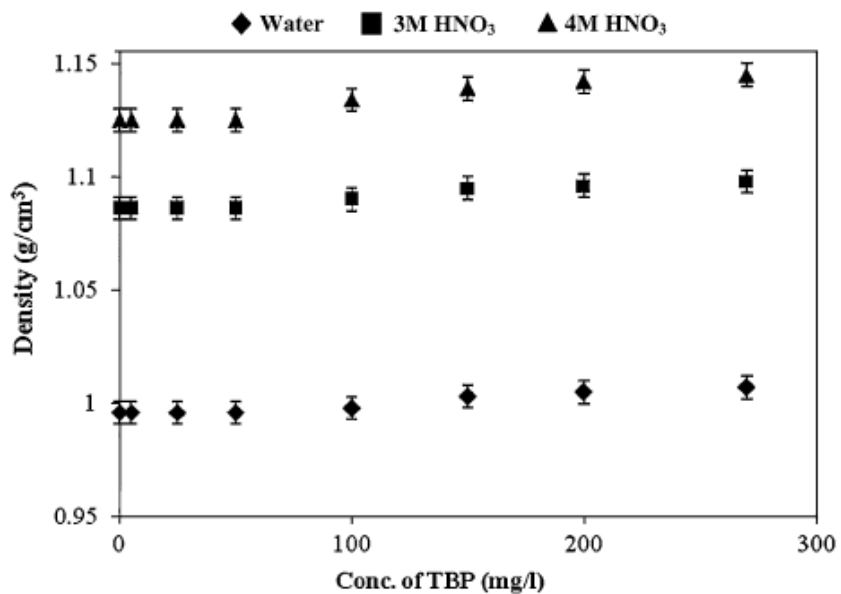

Figure B-15   Effect of TBP on density of the aqueous solutions [Bajoria 2012]
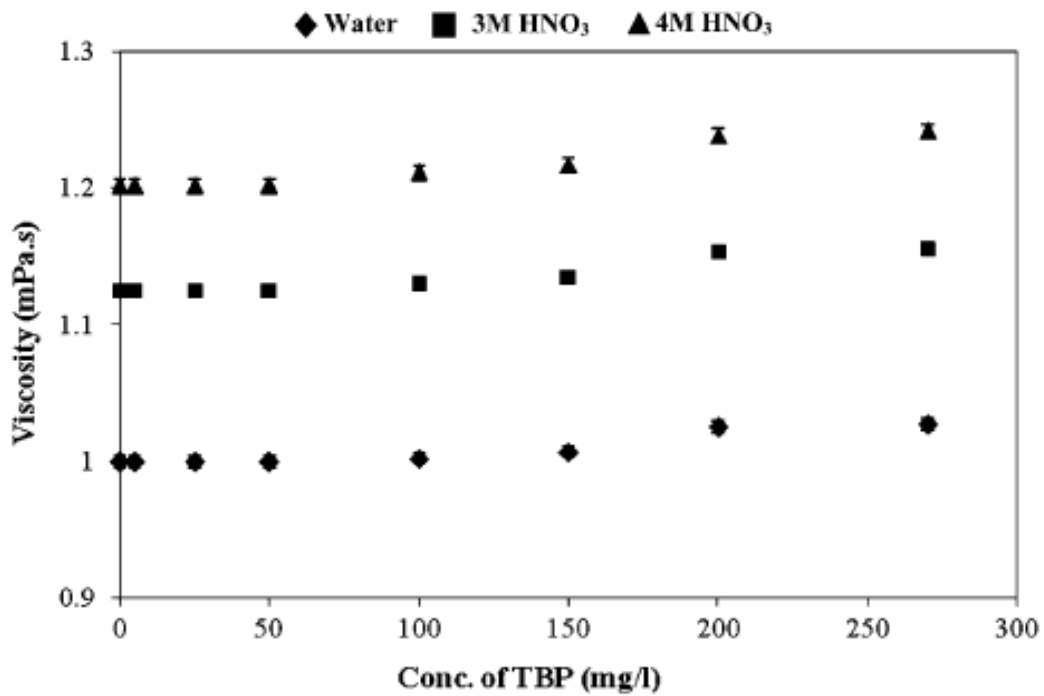
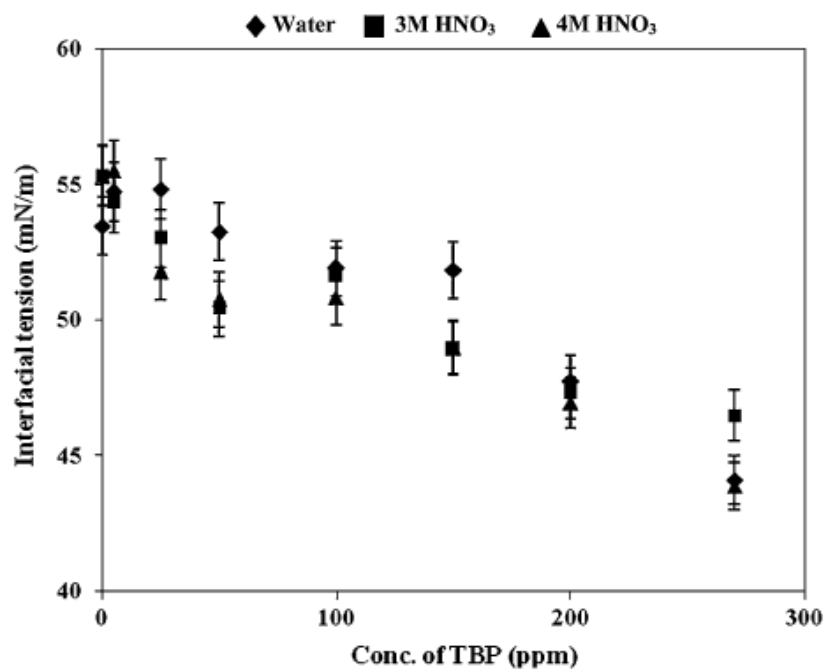**Figure B-16   Effect of TBP on viscosity of the aqueous solutions [Bajoria 2012]**

Figure B-17   Effect of TBP on interfacial tension between dodecane and aqueous solutions [Bajoria 2012]
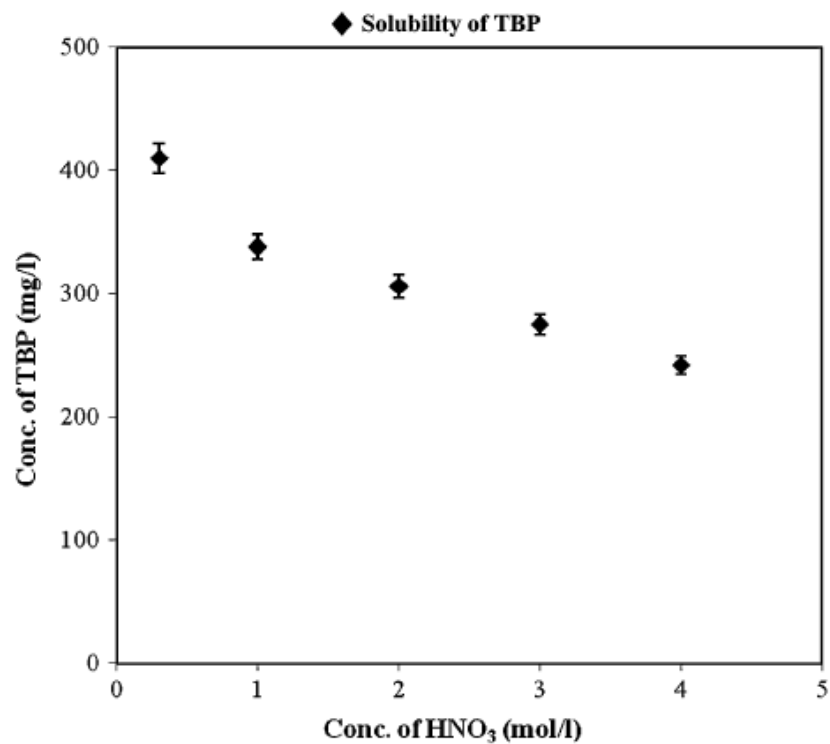
Figure B-18   Effect of nitric acid concentration on the solubility of TBP in the aqueous phase [Bajoria 2012]

Table B-7 Summary of inputs for modeling TOMSK-7 accident

Parameter	Value	Comments
Vessel Dimension		Right cylinder geometry is assumed for simplifying the modeling effort, using data from Table B-5. The properties for this vessel are given in Table B-2.
Inside radius	1.384 m	
Inside height	4.238 m	
Wall thickness	1.6 cm	
Contents		
Liquid density	1.4824 g/cc	It is based on the total contents divided by the total liquid volume as shown in Table B-5.
Liquid volume	25,500 liter	
Gas density	7.29×10^{-4} g/cc	It is assumed that the gas composition followed Eq. B-2, then normalized all gases produced, and evaluated at the average design pressure (12-16 atm) and design temperature of 135 °C [Rodriguez 1995]. It is assumed that ideal gas applied. The average MW of the gas is about 36.14 g/mole.
Gas volume	10,900 liter	See Table B-5.
Fluid Properties		
Viscosity	1.2 mPa-s	Using the top curve of Figure D-5
Surface Tension	45 mN/m	Using the lowest data point in Figure D-6
Explosion		It is assumed that the explosion is due to the decomposition of TBP and the subsequent reactions of the butanol
Energy Generated	1.0×10^{10} J	Using the lower end reported value from Eq. B-1, accounting for the uncertainty of the reaction zone before the explosion.
Gas generated	90 m^3	It is assumed that the calculated gas volume of 100 m^3 minus the initial gas of > 10 m^3 as shown in Table B-5. The gas release rate should be corresponding to the explosion rate modeled in the simulation.

B.4 Tank A-109 HAN-Nitric Acid Explosion (1997)

A hydroxylamine nitrate (HAN) explosion had occurred on May 14, 1997 in Tank A-109 in Room 400 of the Plutonium Reclamation Facility (PRF), which is a part of the Plutonium Finishing Plant (PFP) located in the 200 West Area of the Hanford Site [DOE 1997]. Figure B-19 shows the schematic layout of the fourth floor for the PRF, where Tank A-109 is located. As shown in this figure, this floor houses Operating Control Room 44, Glovebox Room 43, Miscellaneous Treatment Room 41, and Column Room 42 in which vertical sections of two liquid-extraction columns (housed in a glovebox) penetrate the room from above and below. A-109 is one of the chemical makeup vessels that are located in Chemical Preparation Room 40 (see Figure B-19). This room does not contain radioactive materials. The facility ventilation system is designed for once-through flow to allow outside air to flow from uncontaminated areas to contaminated areas, then through a high-efficiency filter system before being exhausted by fans to the atmosphere via the PFP's main stack. An alarmed monitor that was capable of detecting NO and NO$_2$ gases in Room 40 had been installed. Figure B-20 shows the Schematic of the Tank A-109 and its components. As shown in this figure, Tank A-109 was equipped with a motorized agitator that was attached to the outside of the tank's lid. Both the agitator shaft and mixing propellers were suspended from the tank lid to mix the solution inside the tank. The lid was attached with a large number of steel bolts. This tank has a 2" sloped bottom to ensure that the tank contents drain completely. Temperature indication capability is not equipped for this tank. This tank is linked to the vent header above the tank lid. The header is connected to all tanks in Room 40 through an exhaust duct, which vents to the facility exhaust system and then through the stack. No valve exists on either the overflow line, the vent line or vent header. As shown in

this figure, there is a connection to a hydrazine Tank A-109A. Since hydrazine was no longer used, the connecting tank is emptied. The tank has a capacity of 400 gallons and is used to prepare a HAN and HNO_3 solution.
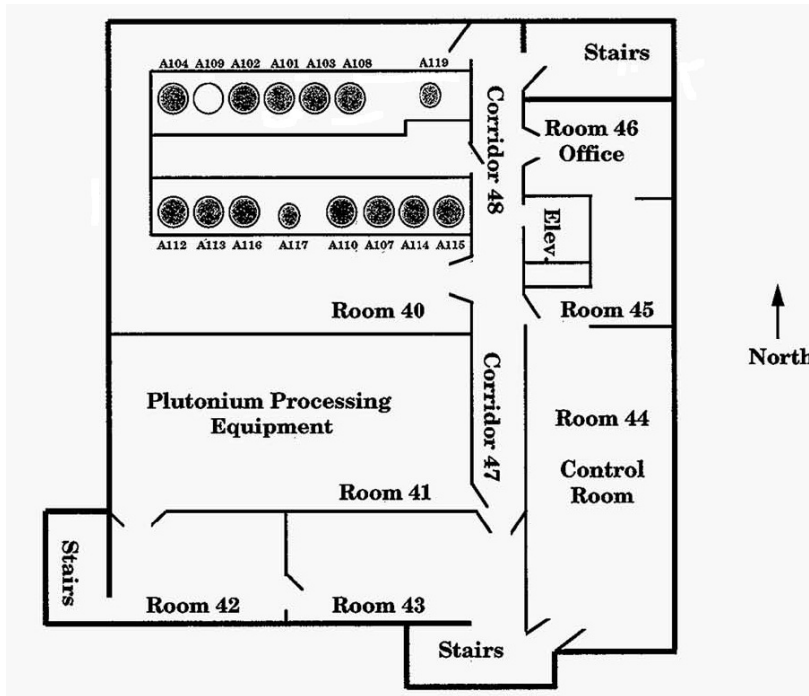

Figure B-19 Schematic of the fourth floor of the PRF and location of Tank A-109 [DOE 1997]
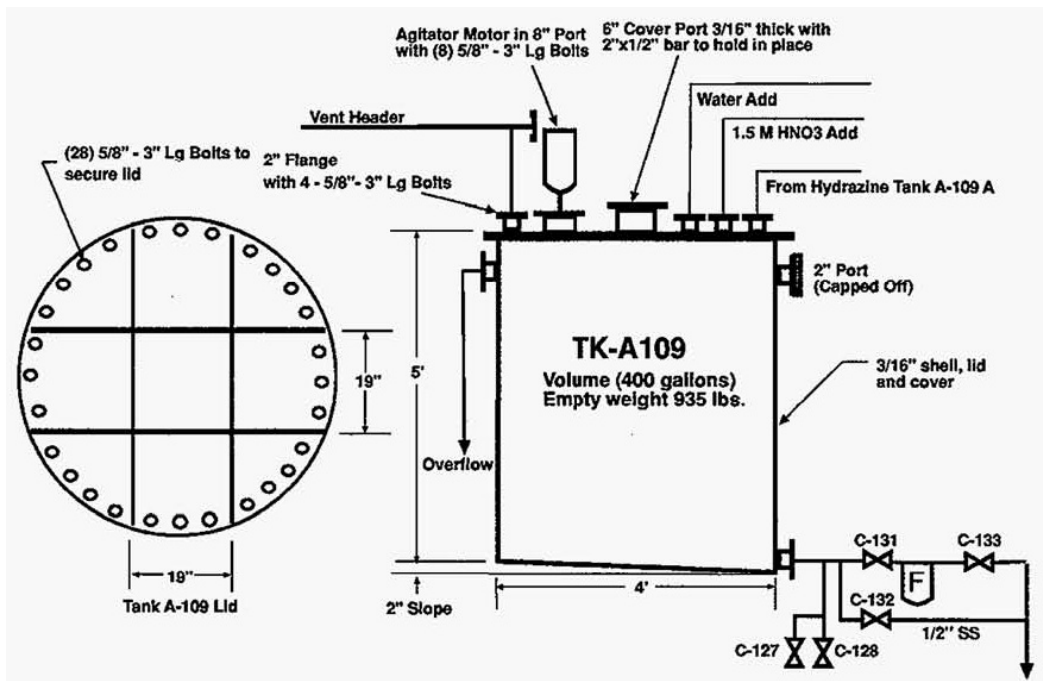

Figure B-20 Schematic of Tank A-109 [DOE 1997]

The sequence of the events that developed the conditions prior to the explosion on May 14, 1997 is given below [DOE 1997]:

- The HAN and NNO_3 solution in the tank had evaporated for nearly four years. It was reported a final sample contained 0.2534 M HNO3 and 0.3542 HAN solution at 370 gallons (3400 lb) before the facility was shut down on June 18, 1993. The inventory in the tank was at 2440 lb on December 29, 1993. The last recording was done in October 28, 1996, which showed a weight of 295 lb (< 30 gallons) remained in the tank.

- The evaporation process, which concentrated the HAN and HNO_3 and the potential effect of catalysts (such as iron from erosion of the tank wall) created the conditions that led to an autocatalytic chemical reaction.

- The explosion inside the tank tore the lid from the tank and propelled it and its attachments upward with enough force to sever a 1.5" fire-suppression water line and to severely damage the ceiling and roof. At some point, the agitator motor broke loose from the tank lid and landed on the floor, while the agitator shaft became embedded in the ceiling. The tank was displaced from the scale on which it was set and came to rest at an angle against Tank A-102, which has been displaced 6" without damaging A-102. The impact of the objects hitting the ceiling, creating a 2-foot bulge in the roof above Room 40, 6-foot-long separation at the roof-wall interface, and a single tear near the bulge. A small crack was observed in the insulation material of a ventilation supply duct on the facility roof. (See Figure B-21 to Figure B-24 for the post-explosion conditions of the facility.)

- Estimating the explosion due to the gas pressure inside of A-109 that blew the tank's lid and displaced the tank [DOE 1997]:

  o Fauske and Associates estimated 200 to 300 psi of pressure with concentrated HAN > 8 M.

  o Babcock and Wilcox Alliance Research Center estimated a pressure buildup of 150 to 250 psi would be sufficient to cause this type of failure to the tank.

Figure B-21   A post-explosion photo of the tanks inside Room 40 and the location of agitator motor [Harlow 1998]

Figure B-22   A post-explosion photo showing the displacement of A-109 Tank [Halow 1998]
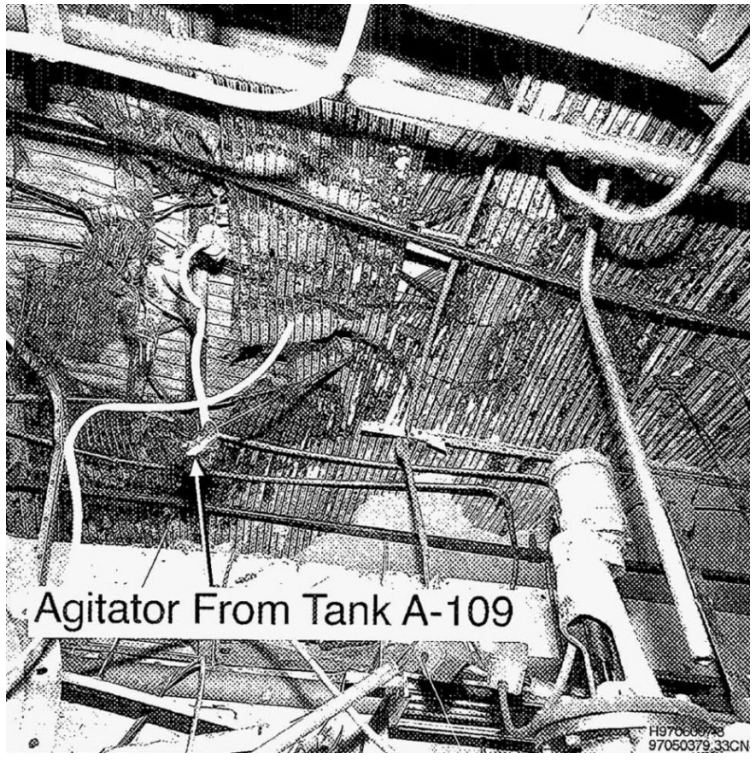
Figure B-23   A post-explosion photo showing the agitator lodging into the roofing area [DOE 1997]
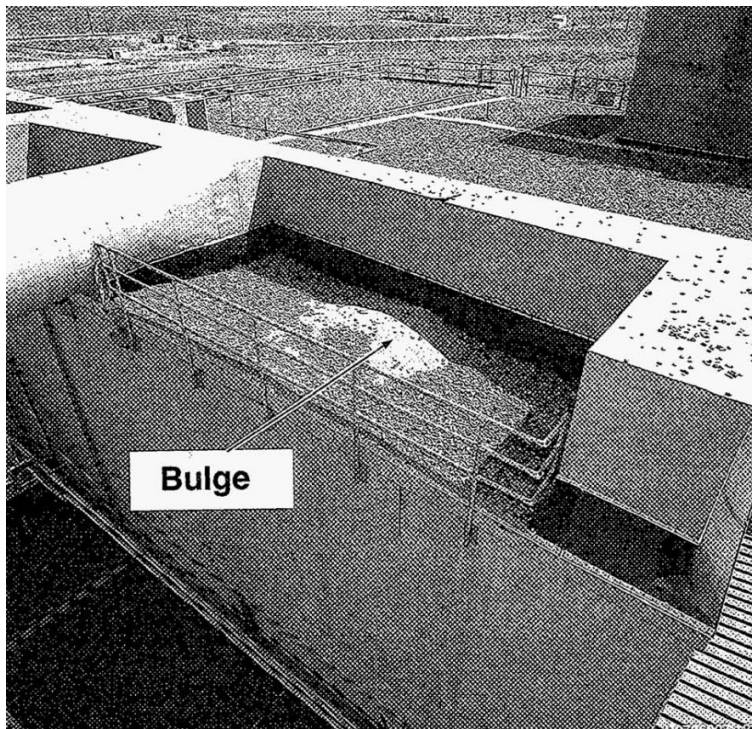

Figure B-24   A post-explosion photo showing the roof bulging [DOE 1997]

Using the pressure data given, the estimation of the explosion energy is calculated using the volume of the tank (400 gallons or 1.51 m^3):

- Using 200 psi (1.38x10^6 Pa) and the tank volume, the explosion energy is 2.08x10^6 Joules.

- Using 250 psi (1.72x10^6 Pa) and the tank volume, the explosion energy is 2.60x10^6 Joules.

If using the last recorded and sampled inventory before shutdown on June 18, 1993, the HAN loss was assumed due to the transfer of 1000 lb out before December 29, 1993. The following calculation of the HAN decomposition energy can be calculated:

1. Last recorded composition of 0.2534 M HNO$_3$ and 0.3542 M HAN solution of 3400 lb (1542 kg) and a volume of 370 gallons.

2. 370 gallons equal to 1400.6 liters in volume.

3. HAN quantities are:

    a. Molecular weight of HAN is ~96 g/mole, assuming that chemical formula of NH$_2$OH•HNO$_3$.

    b. Using the volume from 2 and 0.3542 M HAN from 1 above, 496.09 moles of HAN are calculated. Adjusting the 1000 lb loss after the recorded sample, then this quantity is reduced to 350 moles.

    c. Using the molecular weight given, there is 47644.7 g of HAN. Adjusting the 1000 lb, this quantity is reduced to 33632 g.

4. HNO$_3$ quantities re:

    a. Molecular weight of HNO$_3$ is ~63 g/mole

    b. Using the volume from 2 and 0.2534 M HNO$_3$ from 1 above, 354.91 moles of HNO3 is calculated. After adjusting a 1000 lb loss, this quantity is reduced to 251 moles.

    c. Using the molecular weight given, there is 22359.46 g of HNO$_3$. After adjusting a 1000 lb loss, this quantity is reduced to 15788 g.

5. According to DOE/EH-0555 [Harlow 1998], it gives a number of decomposition energy values, depending on the reactions and concentration of HAN in the HNO$_3$ solution:

    a. For 5NH$_2$OH•HNO$_3$ → 3N$_2$ + 8H$_2$O + 4HNO$_3$, decomposition energy is -68.3x10^3 cal/mole.

    b. For 4NH$_2$OH•HNO$_3$ → 2N$_2$O + 7H$_2$O + 2HNO$_3$, decomposition energy is -49.1x10^3 cal/mole.

    c. For a 0.5 M HAN solution, decomposition energy of 34.1 cal/g of solution.

6.   Using the adjusted mole value for the HAN in 3 above, the decomposition energy according to the first two values in 5 above are:

      a.   21 MJ for $5NH_2OH \cdot HNO_3 \rightarrow 3N_2 + 8H_2O + 4HNO_3$ reaction
      b.   18 MJ for $4NH_2OH \cdot HNO_3 \rightarrow 2N_2O + 7H_2O + 2HNO_3$ reaction

Using the adjusted mass value for the HNO_3 in 4 above, the decomposition energy according to the last value in 5 above is given as 3.2 MJ for 0.5 M HAN solution with HNO_3.

The properties of the HAN solution are estimated as:

1.   Density of the final solution prior to explosion is calculated based on the remaining volume of <30 gallons (114 liters) and 295 lb (133.81 kg):

      a.   Specific gravity (SG) of 1.08518 for HAN and 1.3795 for HNO3 at 25 C assumed [9]

      b.   Assuming HAN mass of 33.6 kg remained, the total volume occupied by HAN is 31 liters, using the SG given in a above.

      c.   Assuming HNO_3 mass of 15.8 kg remained, the total volume occupied by HNO_3 is 11.45 liters using the SG given in a above.

      d.   The remaining mass is assumed occupied by water which is 133.81 kg – 33.6 kg – 15.8 kg is 84.41 kg. Assuming SG of 1 for water, this mass occupies a volume of 84.41 liters.

      e.   Then, the total volume of HAN, HNO_3 and water is 126.86 liters, which exceeds the value given above. Therefore, the water mass is adjusted to be 114 – 31 – 11.45 liters = 71.55 liters. Thus the water mass is 71.55 kg.

      f.   Therefore, the density of the solution is 1.06 g/cc.

2.   Using the final masses of HNO_3 and HAN computed from 1 above, the solution is 3.42 M HAN and 15.3 M HNO_3. In terms of mass ratio, this solution is 25.5wt% HAN and 74.5wt% HNO_3. According to Ref. [10], the reactions at higher concentrations (> 6 M HAN) result in the complete vaporization of the solution.

3.   It is assumed that all HAN is consumed during the explosion, therefore, the remained solution is HNO_3 and water. The properties for HNO_3 are used for aerosol calculations.

The input model for this accident is given in Table B-8.

Table B-8   Model input for Tank A-109 explosion

Parameter	Values
Tank volume and dimension	
Volume	400 gallons (1.51 m³)
Height x Diameter	5 ft (1.52 m) x 4 ft (1.22 m)
Wall thickness	3/16-inch (4.76x10⁻³ m)
Material	347 stainless steel*
Solution (mainly HNO3 after explosion)	30 gallons (0.11 m³)
Gas headspace	370 gallons (1.4 m³)
Solution	
Density	1.06 g/cc
Viscosity$	0.749 mPa-s
Surface Tension$	41.2 mN/m
Explosion Energy	
Pressure-Volume Relation	2.08-2.6 MJ
Decomposition Energy	3.2-21 MJ
Placement of Explosive	Above the solution

*Using properties as given before for 304 stainless steel

$These values are for HNO_3. HAN values are 1 mPa-s and 40 mN/m, respectively [Vosen 1990]

B.5   Additional Possible Vessel Geometries for Considering in Explosions

The process vessels used in spent fuel reprocessing can be classified into two types: unheated and heated [Lux 1995]. The unheated process vessels are the mixer-settlers, holding tanks and storage tanks that do not be externally heated. The heated process vessels, which need external heating are the evaporators, denitrators, and concentrators. External heat used include propane, natural gas, and steam. In both types, organics (TBP or HAN) mat be present in the vessels. Although the accident frequency for the unheated vessel is generally lower than the heated vessels, the possible chemical explosion does exist. The heated process vessels requires accurate monitoring and controlling of the solution temperature and pressure within the vessel to reduce any potential explosions due to chemical reactions and overpressure.

This section describes the possible vessel geometries in the operations that could potentially have an explosion in a spent fuel reprocessing facility.

Unheated process vessels used in the spent fuel reprocessing facilities include mixer-settlers, holding tanks, and storage tanks. Mixer-settlers (see Figure B-25) are used in the extraction operations of the reprocessing, which separates out the aqueous and organic streams. There are also two other process vessels that are used similarly as mixer-settlers: pulse columns (see Figure B-26) and centrifugal contactors (see Figure B-27).
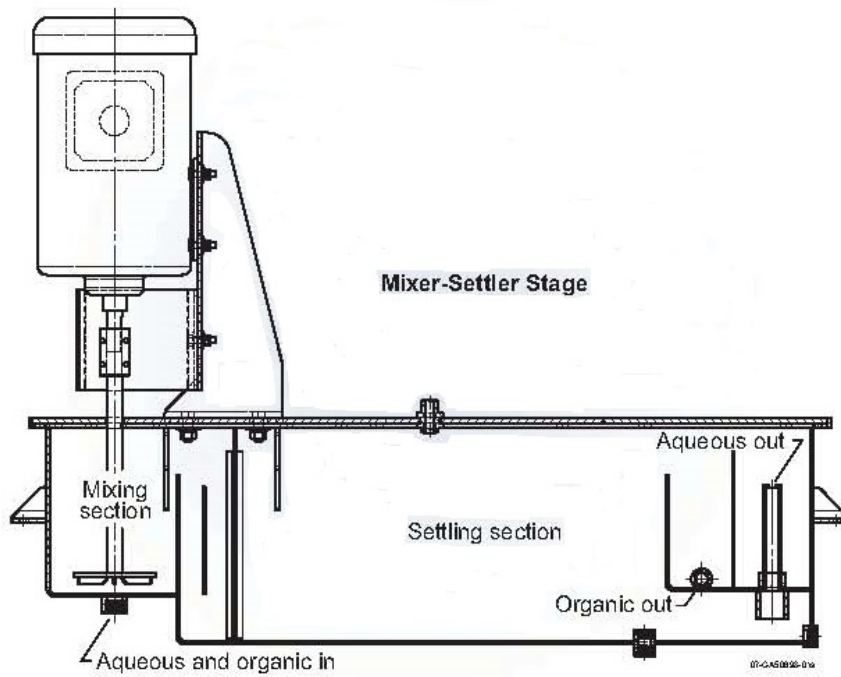
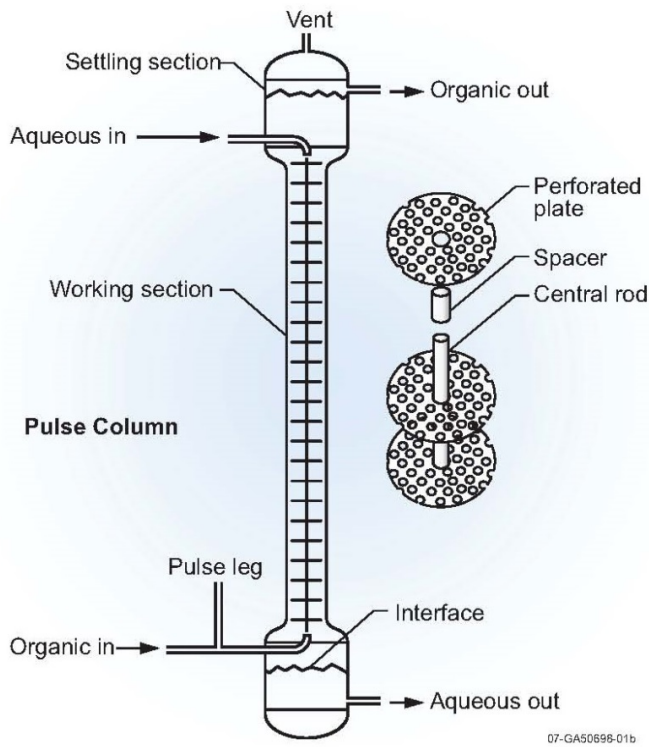**Figure B-25   Mixer-settler schematic [Simpson 2010]**

Figure B-26   Schematic of Pulse Colum with Perforated Concept [Simpson 2010]
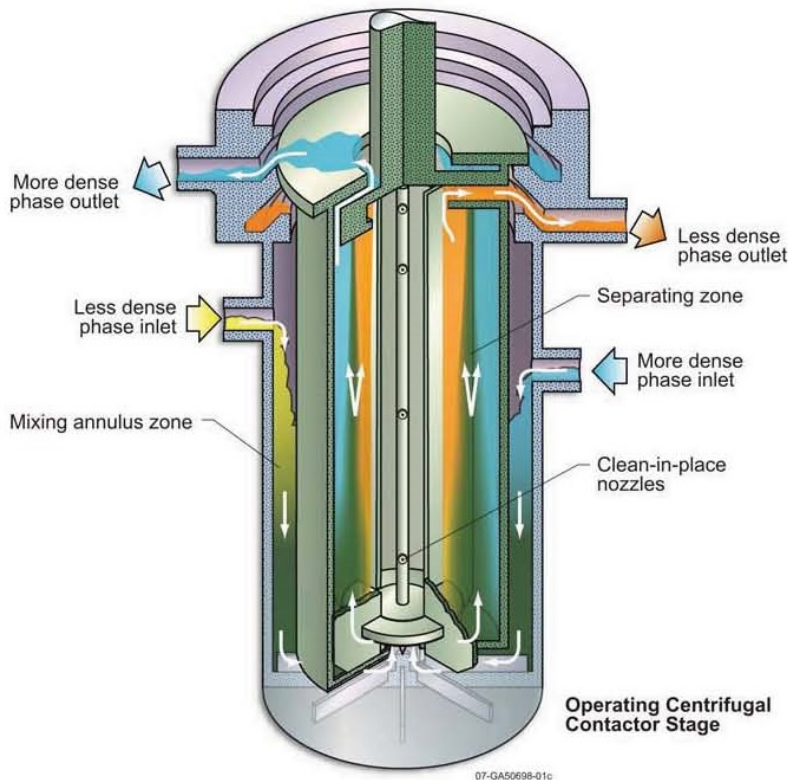
Figure B-27   Schematic of a Centrifugal Contactor [Simpson 2010]

Mixer-Settlers: This equipment provides a single stage of extraction. As shown in Figure B-25, both mixtures enter the mixing section of the equipment. After mixing, the solution flows into the settling section where the separation of the phases takes place. The principal behind the separation is based on the density difference of the phases, using gravity effect. Usually the lighter material (organic) flows over a weir; whereas, the heavier material (aqueous solution) flows out near the bottom of the settling section. The residence time takes from several to tens of minutes, and requires a large facility footprint, but not much headspace and only limited remote maintenance capability because of the nature of the process.

Centrifugal contactors: This equipment is similar to the mixer-settlers, with a single stage of extraction. As shown in Figure B-27, the mixing of the less and more dense phase of fluid is located in the annular region of the contactor. Once mixed, the solution enters at the bottom of the rotor located at the center, where the centrifugal force separates the denser and lighter solutions. The use of rotor to separate the phases is the main difference between this and the mixer-settler. This equipment uses a residence time and requires a small facility footprint and minimal headspace. However, the use of the rotor requires remote maintenance capability for the motor and/or rotor. A cut-away view of the contactor is shown in Figure B-28. This contactor is being used at La Hague plant. Similarly, this type of contactor was to be used at the Barnwell Nuclear Fuel Plant (see Figure B-29). This contactor was used in the first extraction cycle for separating fission products and uranium-plutonium for the head activity separation. This contactor is a stacked 10-stage contactor, which takes in the aqueous feed consisting of ~70% from the adjusted dissolver product from the HA feed tank, and ~30% from the HS column raffinate. The output from this contactor consists of two streams: uranium and plutonium and 30% TBP solution, and fission product aqueous waste stream.

Figure B-28   Cut away view of a multi-stage centrifugal contactor [Drain 2003]
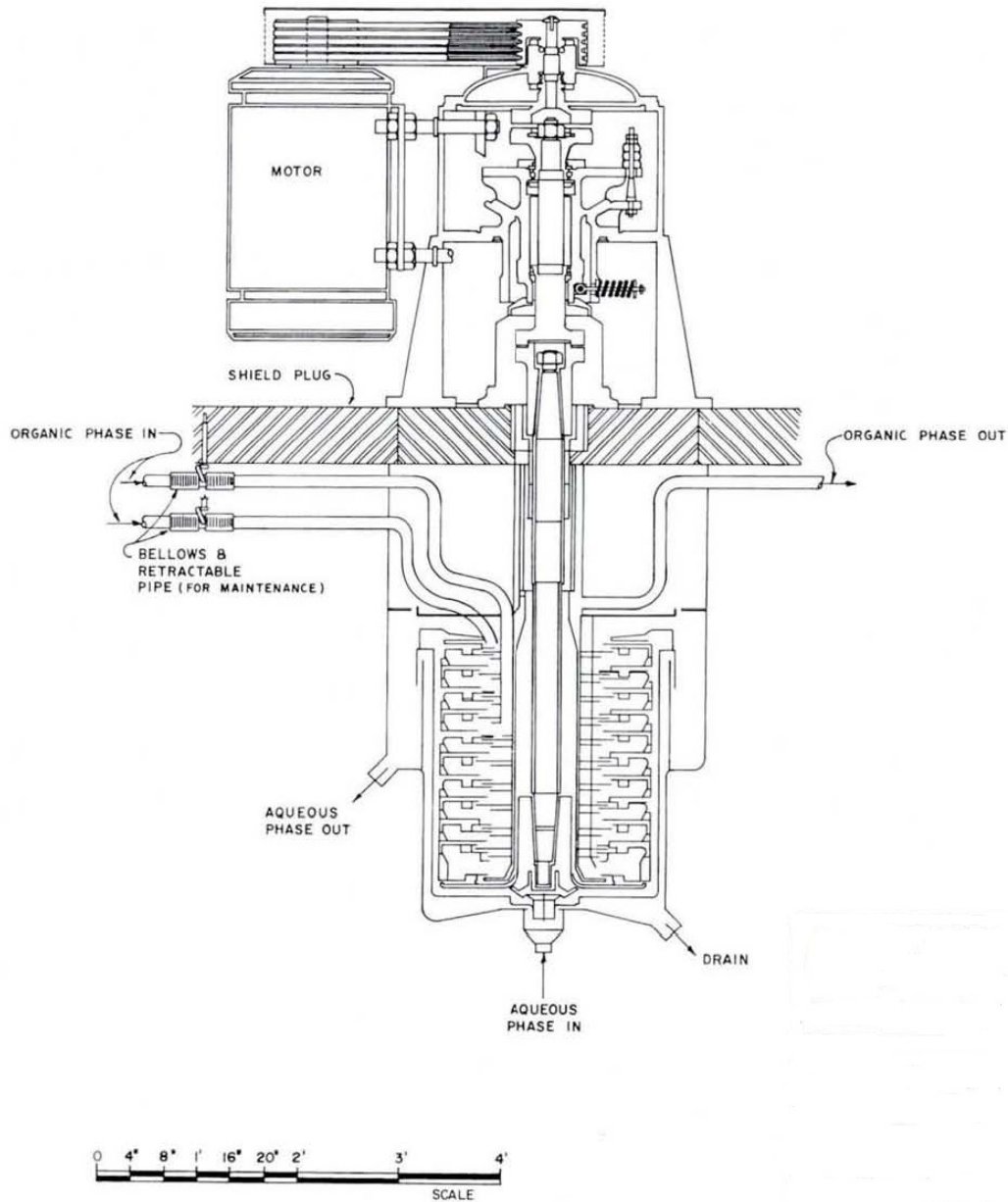
Figure B-29   Cross Sectional View of High Activity Centrifugal Contactor at Barnwell Nuclear Fuel Plant (see Appendix A)

In terms of heated process vessels, concentrators, evaporators and denitrators are used to concentrate the product stream and waste stream to reduce the water or liquid contents. Solidification processes may be followed once these contents are reduced. Examples of the evaporator and denitrators can be found in Appendix C.

Both Appendix A and Bader [2011] provide a number of unheated and heated process vessels that are used in the design of the spent fuel reprocessing facilities. Readers are encouraged to review these appendices for the examples of the process vessels:

Barnwell Nuclear Fuel Plant (Appendix A):

- General Purpose Concentrator
- Acid Fractionater, reboiler and condenser (steam is heated fluid)
- Solvent burner and quench pot
- UPC Concentrator with condenser and reboiler
- PPC concentrator and reboiler

Engineered Alternative Studies [Bader 2011]:

- UREX Process Train – Off-specification evaporator (heated by steam)
- U/Tc Separation Train
  - Pyrolysis oven for removing nitrates from the spent resin, and
  - Carbon reforming oven to remove carbon from the resin.
- CCD-PEG Train – Raffinate evaporators – 40% reduction in volume
- U Solidification Train
  - U hydrate evaporators to concentrate uranium solution
  - U denitrators/reactors (electrical heated) to form UO3
- U/TRU Solidification Train – U/TRU calciner
- Fission Product Solidification Train – calciners (electrical heated)
- Cs/Sr Solidification – steam reformers

B.6 References

[ASM 2012]   Data sheet from http://asm.matweb.com for AISI Type 304 Stainless
                  Steel, July 3, 2012.

[Bajoria 2012]   S.L. Bajoria et al., "Effect of tri-n-butyl phosphate on physical properties of
                  dodecane-nitric acid system," *J Radioanal Nucl Chem*, DOI
                  10.1007/s10967-012-1832-9, May 25, 2012.

[Brennan 1951]   **KAPL-M-MEB-1**, M.E. Brennan, "Physical and Chemical Properties of
                  TBP," Knolls Atomic Power Laboratory, March 9, 1951.

[Colven 1953]   T.J. Colven, Jr. et al., Interim Technical Report: TNX Evaporator Incident
                  January 12, 1953, E.I. du Pont de Nemours & Co., May 15, 1953.

[Decker 1987]   **BRL-TR-2864**, M.M. Decker et al., "HAN-Based Liquid Gun Propellants:
                  Physical Properties," Technical Report, AD-A195 246, US Army Ballistic
                  Research Laboratory, November 1987.

[DNFSB-T33]   **DNFSB/TECH-33**, "Control of Red Oil Explosions in Defense Nuclear
                  Facilities 2003," Defense Nuclear Facilities Safety Board Technical
                  Report, November 2003.

[DOE 1997]   **DOE/RL-97-59**, Accident Investigation Board Report on the May 14, 1997
                  Chemical Explosion at the Plutonium Reclamation Facility, Hanford Site,
                  Richland, Washington, Revision 0, Department of Energy, July 26, 1997.

[Drain 2003]   F. Drain et al., "40 years of Experience with Liquid-Liquid Extraction
                  Equipment in the Nuclear Industry," Waste Management 2003
                  Conference, February 23-27, 2003, Tucson Arizona, U.S.A.

[Durant 1988]   **Savannah River Laboratory**, W.S. Durant et al., "Explosions and Other
                  Uncontrolled Chemical Reactions at Non-Reactor Nuclear Facilities of the
                  Savannah River Plant," DP-MS-88-15, E.I. du Pont de Nemours & Co.,
                  December 1988.

[Eisenhawer 1995] **LA-UR-95-1712**, S. W. Eisenhawer, "Accident Reconstruction Using
                  Process Trees," Los Alamos National Laboratory, 1995.

[Hang 1998]   **WSRC-TR-98-00292**, T. Hang, "The FB-Line and F-Canyon HAN/Nitric
                  Acid Decomposition Study," Westinghouse Savannah River Company,
                  August 19, 1998.

[Harlow 1998]   **DOE/EH-0555**, D.G. Harlow et al., "Technical Report on Hydroxylamine
                  Nitrate," Department of Energy, February 1998.

[Harmon 1976]   H.D. Harmon et al., "Behavior of Tributyl Phosphate in A-Line Processes,"
                  DP-1418, E.I. du Pont de Nemours & Company, Savannah River
                  Laboratory, August 1976.

[Hyder 1994]   **WSRC-TR-94-059**, M.L. Hyder, "Safe Conditions for Contacting Nitric Acid or Nitrates with Tri-n-butyl Phosphate (TBP)," Westinghouse Savannah River Company, January 1994.

[Klophenstein 1958] R.K. Klophenstein, Studies of the Behavior of the TBP-Kerosene Solvent in Uranium Refining: Degradation of the Kerosene Diluent, NLCO-769 (TID-4500, 14th Ed.), National Lead Company of Ohio, August 12, 1958.

[Lang 1968]   G.P. Lang et al., "Some Physical Properties of Uranyl Nitrate Solutions – Topical Report," Report No. MCW-1424 (TID-4500, 14th Ed.), Mallinckrodt Chemical Works, St. Louis, December 15, 1968.

[Lux 1995]   **WSRC-RP-95-910**, C.R. Lux and R.E. Vail, "Overview of "Red Oil" Frequency Analyses for F-Canyon (U)," Rev. 0, Westinghouse Savannah River Company, September 21, 1995.

[Mckibben 1976]   J.M. McKibben, "Explosion and Fire in the Uranium Trioxide Production Facilities at the Savannah River Plant on February 12, 1975," DPSPU 76-11-1, E.I. du Pont de Nemours & Company, Savannah River Plant, October 1976.

[Ondrejcin 1961]   R.S. Ondrejcin, "Physical Properties of Uranium Process Solutions," DP-653, E.I. du Pont de Nemours & Co., Savannah River Laboratory, October 1961.

[Paddleford 1994]   **WSRC-MS-94-0649**, D.F. Paddleford and H.K. Fauske, "Safe Venting of 'Red Oil' Runaway Reactions," Westinghouse Savannah River Company, December 21, 1994.

[Rodriguez 1995]   **LA-UR-95-1756**, E.A. Rodriguez and R.F. Davidson, "TOMSK-7 Radiochemical Facility Explosion: Chemical Vessel Burst Failure Analysis," Los Alamos National Laboratory, 1995.

[Simpson 2010]   **INL/EXT-10-11753**, M.F. Simpson and J.D. Law, "Nuclear Fuel Reprocessing," Idaho National Laboratory, February 2010.

[Smith 1994]   **WSRC-TR-94-0540**, J.R. Smith and W.S. Cavin, "Isothermal Heat Measurement of TBP-Nitric Acid Solutions," Westinghouse Savannah River Company, December 16, 1994.

[Usachev 2003]   V.N. Usachev and G.S. Markov, "Incidents Caused by Red Oil Phenomena at Semi-Scale and Industrial Radiochemical Units," *Radiochemistry*, Volume 45, No. 1, 2003.

[Vosen 1990]   S.R. Vosen, "Hydroxylammonium Nitrate – Based Liquid Propellant Combustion – Interpretation of Strand Burner Data and the Laminar Burning Velocity," *Combustion and Flame*, 82 (1990), pg 376-388.

APPENDIX C

MISCELLANEOUS DATA

C.1   Program Review Comment Responses Given February 7, 2013

**Comments on Sandia National Laboratory Research Program Review February 7, 2013
Source Term Releases from Spent Nuclear Fuel Reprocessing Facilities, JCN V6050**

1.   Ensure the aerosol modeling incorporates current aerosol physics and chemistry to describe aerosol transport and behavior during all releases.

      We plan to use best-estimate aerosol and phase modeling to describe transport and releases into the environment.

2.   The radionuclide source term releases should be expressed in radioactivity units of Ci/day, Ci/month, etc. and concentrations in releases to the atmosphere and aqueous water bodies, including groundwater, where appropriate. Exposure of radioactive releases needs to consider both workers and members of the public (e.g., reasonable maximally exposed individual).

      The modeling approach that we envision will treat the mass of radionuclides in various locations, including suspended particles (aerosols). The radionuclides will be assumed to be distributed uniformly in the masses that are tracked, so the mass and activity fractions are identical. Thus, at the end of the analysis, releases in terms of isotopic activities are readily calculated by the tool and provided to the user.

3.   Clarify how fires will be treated empirically and how fires will be modeled.

      Codes such as MELCOR are not designed to model fire physics explicitly. The fire can be approximately modeled in MELCOR and other similar codes by adding the fire mass (such as soot/by-product) and fire energy into the room(s) or control volume(s). An empirical correlation is mainly involved with the development of the mass and energy source profile(s) that can be input to codes, such as MELCOR. These empirical correlations (temperature profile in the accident room) may be developed using codes specifically designed for fire analysis, such as FDS or CFAST. The correlation is developed using this approach for a range of the fire size (in energy) and room size, etc. Ultimately the developed source term tool should be able to be benchmark against "defined" fire experiments that were used to validate the fire analysis codes. Depending on the resources available, it may be worthwhile to modify codes, such as MELCOR, to be able to model fire scenarios, without relying on the empirical correlation approach. In fire analysis codes, the atmosphere in the room can be modeled with two gas layers (hot layer and cooler layer). The height of the hot layer is adjustable depending on the fire energy added to the room. For a code such as MELCOR, hot gas layer modeling cannot be performed.   Rather, the modeling approach would rely on the user to divide the room into appropriate, smaller control volumes in order to capture the stratification effect. We currently intend to use the correlational approach when developing the characterization of fires within the source term tool.

4.   For the major process, include the off-gas treatment processes, red-oil mechanisms, calcination and oxalate processes during the uranium/plutonium purification processes.

It is our intention to cover these processes, including the vitrification process, and for the range of accident conditions. Below is a table (Table C-1) from the DP-1558 report. We intend to add additional scenarios as needed to cover all known processes and accidents in the literature we identified.

Table C-1 Potential incidents possible at a spent fuel reprocessing facility [DP-1558]

Low Consequence	Intermediate Consequence	High Consequence
Fuel Receiving and Storage Operation		
• Fuel damage in transit or upon arrival • Fuel cooling time too short • Loss of cooling • Surface contamination not removed from cask • Low water level • Rupture of fuel during storage	• Cask inadvertently vented • Cask dropped • Damage to fuel assembly outside cask • Damage to stored fuel • Loss of cooling capability	• Hydrogen explosion in fuel cleaning vessel • Criticality
Shearing Operation		
• Fuel jammed or stranded in shear • Irregular length of hulls • Shear jammed	• Malfunction of inert gas system • Chopped fuel overheating • Pressurization of shear • Fuel element overheating	• Pyrophoric fire[1] • Release of volatile and particulate activity into room
Dissolving Operation		
• Un-complex fluoride in dissolvent • Pu-rich reside settling in dissolver, lines, and other process vessels • Dissolver seal failure • Inadequate cladding rinse • Transfer error of dissolver solution • Suckback (due to eructation and pressurization of dissolver) • siphoning	• Contact of sheared fuel at a temperature about 300 ˚C above boiling point of dissolver solution • Pressurization of the dissolver • Dissolver leakage (due to corrosion gasket/valve failure) • Charge of inadequate cooled fuel • Malfunction of dissolver off-gas (VOG) iodine adsorbers • Excessive interaction of fuel external to dissolver (slightly reduction in criticality safety margin) • Excessive pressure in water wash compartment except during basket exchange. • Dissolver pot coils not submerged during shutdown (high release of volatiles to room atmosphere) •	• Precipitation of Pu polymer (potential criticality) • Zirconium fire[1] • Zirconium explosion (due to significant fines in the dissolver)[1] • Explosion in the iodine adsorber • Overconcentration in dissolver (potential criticality) • Self-concentration of dissolver solution (potential criticality) • Low H^+/Pu ratio in dissolver solution (potential criticality) • Inadequate poison in dissolver or leach solution (potential criticality) • High fuel loss in leached fuel fragments • Failure of cladding monitor (potential criticality) • Absence of basket in dissolver • Explosion in dissolver vessel or hold tanks (H_2 explosion)
Solvent Extraction Operation		
• Plutonium concentration in a solvent extraction contractor increases beyond normal values • Potential backup of radioactivity by air lines	• Potential for abnormal accumulation of fissile material by plutonium reflux in contractor, particularly during high activity waste rework (decreased criticality safety margin)	• Solvent fire[2] • Potential for buildup of unsafe amounts of fissile material in organic storage tank (potential criticality) • Potentially unsafe feed concentration (potential criticality)

Table C-1 Potential incidents possible at a spent fuel reprocessing facility [DP-1558]

Low Consequence	Intermediate Consequence	High Consequence
• Flow reduction or interruption of scrub streams to contactors (uranium) • Loss of organic flow to a contactor (uranium) • Low temperature in scrub stream to contactor (uranium) • Low temperature in contactor • Emulsion in solvent washer • High plutonium losses in waste streams from contactors (in PUREX process) • High plutonium concentration in contactor (in PUREX process) • Excess gamma activity in plutonium product	• Potential for excessive product loss to spent organic effluent (reduced criticality safety margin) • Accumulated solvent in rooms • Solvent extraction system contents removed by overflow (in sump)	• Potential for excessive plutonium loss to aqueous waste (potential criticality) • Incorrect material transfer of feed chemicals, product materials or incorrect materials (potential criticality, excessive radioactive material release)
Product Evaporation		
• Leaks in tank containing concentrated uranium-plutonium solutions	• Transfer error in a product evaporator system • Coil or tube-bundle (in reboilers and condensers) failure • Overflow of an evaporator	• Potential reaction between TBP and UNH (red oil explosion) • Overconcentration of U/Pu product (potential criticality) • Boil-over (release of radioactive materials from primary containment)
Waste Evaporation		
• Overconcentration of waste	• Ruthenium escapes to stack • Leaks (to sumps) • Transfer error in the evaporator system • Coil or tube-bundle (in reboilers and condensers) failure • Overflow of a feed or bottom tank • High radioactivity in high-activity waste r evaporator condenser • Inadequate vessel cooling capacity (that leads to release aerosols to room atmosphere. • Leak of high-level waste concentrate	• Boil-over (release of radioactive materials from primary containment) • Explosion in high-activity waste evaporator
Acid Recovery Operation		
• Pressurization of acid absorber/fractionator	• Leaks due to corrosion • Eructation due to foreign materials in feed • Solvent addition to feed tank	• Explosion due to solvent in feed or temperature > 140 ˚C • High radioactivity in recycled acid and/or water
Recovery Ion Exchanger Operation		
	• Overflow • Leakage due to corrosion	• Uncontrolled reaction between nitric acid and anion exchange resin in the primary recovery column • Ion exchange resin fire (lead to radioactivity release to environment)
Off-Gas Treatment Operation		

Table C-1 Potential incidents possible at a spent fuel reprocessing facility [DP-1558]

Low Consequence	Intermediate Consequence	High Consequence
• Loss of off-gas header volume • Iodine removal inadequate (increase iodine accumulation in krypton solvent recovery) • Excess nitrogen oxides in VOG iodine adsorber stream	• High radioactive particulate releases to building ventilation filters (increase release to sand filter) • Off-gas heater inadequately heats streams to iodine adsorbers • High ruthenium adsorber bed temperature • High krypton-85 releases	• Filter failure • Process vent system pressurized (leads to release to sand filter and smaller amount to atmosphere
Uranyl Nitrate Operation		
• Overflow from storage tank • High uranium concentration in recovered acid	• Plugging of instrument lines and sensors during evaporation • Steam coil leak in evaporator reboiler • Cooling coil leak due to corrosion • Overflow in concentration system	
Uranium Denitration and Reduction Operation		
• Pressurization of denitrator • High uranium concentration in recovered acid	• Over-pressurization of redactor • Fire in Ammonia dissociator cubicle • Reductor malfunction which leads to release of uo_2 and hydrogen	• Denitrator eructation (leads to fire and explosions) • Hydrogen explosion (hydrogen leaked from reductor)
Uranium Hexafluoride Operation		
• Uranyl nitrate solution leaks • Uranium hexafluoride release • Airborne uranium oxide		• Criticality due to excessive uranium accumulation (high enrichment fuel) • Fire (due to solvent in feed, hydrogen leak and presence of pyrophoric oxide powders)
Co-conversion Process Operation (UO2-PuO2 production)		
• Transfer errors • Process solution leak • Overflows • Chemical addition error	• Pressurization of calciner • Calciner breached from internal corrosion • Calciner breached from impact (crane drop) • Excessive penetration of calcine through the calciner primary filter • Filter system breached • Fire suppression system failure • Uranium in steam condensate or cooling water returns the uranium oxide dissolver • Loss of power	• Ammonium nitrate explosion in calciner • Hydrogen explosion in product storage due to leak in hydrogen delivery system • Hydrogen explosion in Pu Nitrate storage tank due to radiolysis • Uncontrolled reactions • Fire in process room • Hydrogen explosion in cold chemical area
Waste Calcination Operation		
	• Excessive penetration of calcine through sintered metal filters	• High temperature breach of the calciner • Calciner breached from internal corrosion, thermal shock, pressurization and impact • Filter systems breached •
Solid Waste Processing Operation		

Table C-1 Potential incidents possible at a spent fuel reprocessing facility [DP-1558]

Low Consequence	Intermediate Consequence	High Consequence
• Airborne cement dust in grout mixer area • Radioactive contamination in cement preparation area • Failure of contaminated process components • Drums improperly filled (no cement added)	• Violation of stack release guide • Container failure • Power failure	• Fire in fuel hardware fixation area • Drum overflow in fuel hardware fixation area • Excessive fissile material in hulls • Waste container failure after filling • Fire in β-γ waste facility • Criticality potential in α waste
Solidification of Intermediate level liquid waste		
• Transfer error that leads to high activity waste • Overexposure of personnel to radiation • Mixer plugging, which leads to worker exposure	• Airborne activity due to leaks, overflows, foam-out from mixer, suckback, and loss of vessel ventilation (power failure) • Waste container failure due to weld failure, fault in drum, corrosion, or impact	• Fissile material in feed • Uncontrolled reaction in mixer or product container
Vitrified High-Level Waste Storage Operation		
• Contaminated canisters • Water loss from storage pool	• Loss of cooling water and shielding • Canister stress corrosion • Canisters raised above adequate shield level • Canisters dropped during handling	• fire

[1]During the head-end process when the spent fuel rod is being chopper or crushed, magnesium cladded fuel and uranium/plutonium metal fuel are exposed to air, combustion may occur at normal temperatures [OECD 2005]. Zircaloy debris or dust could pose a potential fire or explosion if this material is used for the cladding [IAEA DS360].
[2]N-dodecane in TBP in the PUREX process poses potential combustion conditions [Ikeda 2003].

5.   Much of the description on aerosol behavior seems to be derived from spent fuel type releases where solid materials become vaporized as aerosols. However, for reprocessing spent fuel, the aqueous separation process involves aqueous solutions of nitric acid and organic solvents. Fine powders following heating and evaporation are other major sources of aerosols. For these situations, where aqueous and organic liquids and fine powders may be converted to aerosols, the processes appear to be less understood including the aerosol sizes that are likely to be formed. It is not clear where vapors are distinguished from aerosols during the source term release processes.

The commenter correctly identifies two general processes for creating aerosols: (1) thermal processes that vaporize materials and subsequent condensation of the vapors to generate the aerosol particles (e.g., a tea kettle that creates steam that subsequently condenses to form fog) and (2) mechanical formation of aerosols by explosions or other highly energetic events that generate large pressure waves and induce stresses and accelerations. Transport codes, such as MELCOR, can handle the thermal processes that form aerosols but lack a model for aerosol generation by mechanical processes. The focus of Gelbard's presentation was on mechanical generation of aerosols, specifically creation of liquid aerosols (containing solvent and dissolved or suspended radionuclides). The mechanical processes are simulated using a suite of codes known as the Sierra Codes.

The Sierra codes model the explosion and approximately up to about 10 seconds after the explosion. During this period both gases and particles can escape from the processing equipment. The processes modeled with the Sierra codes are particle formation by the forced rapid ejection and breakup of the liquid by mechanical processes. We simulate this release period by modeling both the gases and particles, with the assumption that all the gases spread throughout the room containing the equipment that has failed. The drops may turn into solid particles by vaporization of volatile liquids. The Sierra models also include liquid drop particles vaporizing.

Subsequent to the release period, MELCOR models any further particle vaporization, condensation, deposition, and transport to other locations. Thus, the output from the Sierra codes is used as input for MELCOR. (MELCOR does not model the initial release period when material is ejected from processing equipment.) The particles, whether they are liquid or solid are called aerosols when suspended, but are no longer considered to be aerosols if deposited on fixed surfaces such as a wall, floor, or even furniture.

6.   Fission products, transuranic nuclides and activated materials should be considered in development of the source term release models.

We agree with this statement and intend that the tool will include each type of radionuclide at the discretion of the user.

7.   For chemical releases, it is our understanding that chemical reactions, or reactive transport models, will not be considered in the source term models.

We agree with this statement, with a few caveats. First we do intend to include phase change. This would be done by introducing an expression for vapor pressure as a function of temperature. Second, empirical fire models convert organic solvents to combustion products and energy. We do not intend to include a general treatment of either thermodynamic equilibrium or reaction kinetics in the model.

8.   The major assumptions listed in the overview presentation appear reasonable, although additions are expected to be needed as we begin to fully understand the processes involved   and when further information becomes available.

We agree.

9.   The equation for the source term, ST = MR x DR x ARF x RF x LPF, is based largely on empirical correlations and may not be applicable to source term releases from reprocessing facilities using aqueous separations. Limitations of this equation need to be recognized for reprocessing. The parameters, data for the parameters, and all assumptions need to be thoroughly examined for application to reprocessing facilities. The proposed mechanistic modeling would seem to be the appropriate way to address reprocessing facility source terms. A deterministic approach may be reasonable, but a probability approach using Monte Carlo approach or Latin Hypercube Sampling would provide a more realistic means of assessing source term releases.

We agree with the assessment of the five-factor equation often used by the DOE, and it is our intention to improve on this traditional approach to the extent that we are able within the scope of the project.

The initial source-term model is being developed as a deterministic tool, but there are a number of parameters needed to run the model. It is for these parameters that a probability approach could be used in the future, but is not planned as part of this work, which is focused on development of the model. Initially, the user of the source-term tool may need to perform sensitivity analyses to determine ranges of results based on reasonable ranges of input parameters. A probabilistic approach based on the deterministic model could be developed as a follow-on effort if this would have value to the NRC.

10.   What is the approach used to model red-oil explosions, oxalate explosions, and potentially hydrogen (from radiolysis effects) explosions?

All explosions are modeled with the Sierra codes. What differentiates the explosions for red-oil, oxalate, or hydrogen are (1) the amount of energy released, (2) the size of the explosive region within the system, (3) the shape of the explosive region (such as confined to a thin layer, or a small puck-shaped region), (4) the amount of other fluids in the system, and (5) the location of the explosive (i.e. where in the processing equipment). The Sierra models can simulate the explosive release of liquids to form particles once these parameters are specified. Parameters associated with the five features listed above will be different for red-oil, oxalate, and hydrogen.

11.   The slide "COEX-like Process – Daily Throughput Diagram" in the presentation-Fuel Reprocessing Facility and Accidents – is a model of an extraction process separating and combining plutonium with uranium to produce oxide powders for storage and further mixed-oxide reactor fuel. Let's call this Co-Extraction Process I. Another figure should be added that more closely follows the COEX^{TM} – AREVA separation process. We will call this figure Co-Extraction Process II. Both concepts should be discussed in terms of the co-extraction process for the aqueous separation of uranium and plutonium. Because we have no license at this time for the construction and operation of a spent fuel reprocessing facility, it is unknown which of the two separation processes may ultimately be submitted. But it is always easy to scale back from the more sophisticated proposed process, Process I, to the lesser complicated process, Process II, than it is to scale up an analysis from Process II to Process I. This needs further discussion.

We agree that both concepts should be included, since they both have a U product stream and U-Pu oxide product stream. The only difference between them is in the 1st and intermediate extraction cycles where U and U/Pu streams are produced. The other emphasis is the fission product separations in terms of the end FP stream(s) desired. Currently, it is our intention to include Process I (as a detail description, including flow diagram, conceptual plant layout, etc., since we have the information) and the description of the conceptual plant in our Task 2 report. If the description of Process II is available, similar to the level of Process I, we may want to include Process II in the write-up; otherwise, we may include it as a process description. The Barnwell plant has been included in the write-up at present.

12.   The presentation Fuel Reprocessing Facility and Accidents appears to discuss the important aspects of an aqueous separation process for the co-extraction of uranium and plutonium, and provide background on the history of chemical explosions, major accidents at reprocessing facilities, and the role of engineered safety features. Some editorial comments regarding the presentation are:

a. The color coding of Slide 2 is not clearly stated on the slide. First-time readers may have some difficulty in understanding the color scheme. Also some boxes in the separation diagram are not colored.

b. Slide 5 should have columns for "Ci/MTHM" for the major radionuclides.

c. Slide 5: Indicate initial enrichment percent of U-235 (e.g., 5%).

d. Indicate color scheme for Slide 6.

e. In upper right hand corner indicate U-235 enrichment percent.

f. Be aware that this slide provides a separation for all of the TRUs that will be combined with uranium to form the oxide fuel, while Process II (Comment 11 above) only provides for uranium and plutonium to be separated together. The remaining TRUs will be combined with the fission products and vitrified in glass for ultimate disposal as HLW.

g. In Slides 8-11 somewhere, indicated the plutonium oxalate problems that occurred at the Russian facilities. May want to reference some of the papers published by AREVA regarding the plutonium and uranium oxalate separation issues.

h. Spell out names for acronyms when the first appear (e.g., HAN, JCO)

i. Provide list of chemical and expected concentrations that may be expected to be used in the separation processes.

j. Slide 21: Why is nuclear Excursion Accident type listed as a minor public severity for source term releases from reprocessing facilities? The analyses have not yet been performed for reprocessing facilities so one cannot predetermine a severity index for both workers and the public

k. Back-up slides are instructive.

These corrections are noted and will be incorporated into the final report for Task 2.

13. We may wish to select particular processes and/or vessel configurations that should be included in future computational aerosol dynamics simulations of airborne releases from detonations (F. Gelbard's presentation). There are a number of different types of vessel configurations that are incorporated in the reprocessing separation processes. Among them are vessels for centrifugal separation, liquid and solid storage, centrifuge operations, evaporation processes, calcination heaters, etc. These need to be considered.

The plan is to compute releases for different vessels using the detailed code (in Sandia's Sierra code suite) that produced the movie shown at the meeting. Only for demonstration purposes did we use a simple cylinder. The Sierra codes will be used for developing particle release correlations for the different vessels, and these correlations will be incorporated into the tool provided to the NRC. As discussed below for comment #21, the Sierra code suite will not be part of the tool.

14. Similarly, we need further discussion of the direction on the "prototype" plant design that will be used to establish default parameters (Slide 5 of Synopsis presentation).

We agree. It would seem desirable for Sandia to work more closely with those who are developing the prototype plant design so we can better understand evolutions in the prototype design.

15.   We need to ensure that SNL has captured all of the possible accidents that could occur at a reprocessing facility since the modeling is going to be responsive rather than predictive. With that, there is a need to address the off-gas system and consider possible accidents. If one takes a look at the diagram on page 6 of David Louie's handout (Fuel Reprocessing Facility and Accidents), in particular the off gases coming from the fuel shearing/voloxidation box, Ru gas is a major component. Ru is not a gas that we have previously considered. Will this be stored temporarily on site until it has decayed? It is a beta emitter with a half-life of approximately 374 days. It decays to Rh-106, which in turn immediately decays (~ 30 s) to Pd-106 and emits intense gamma rays at 512 keV and 622 keV, which could cause a problem with shielding. Also, they need to consider the cryogenic storage of Kr-85 and its release if the cooling system fails.

It is our intention to cover all anticipated scenarios for the spent fuel reprocessing plants in the report (see comment 4 response). Note that the source term tool we are developing is general and relies on the user to describe the accident simulated. To evaluate if the shielding is adequate or not for Ru decay and subsequent decay products, the user may need to treat this issue outside the source term tool, which is not intended to evaluate radiation shielding.

16.   Following on from a comment in the meeting about considering iodine in the waste solvent. Iodine is likely to be in the solvent at very low concentrations; in the order of ppm. Therefore, Nate was right and we don't need to consider it in terms of release during an accident. Isn't ^{129}I a major radionuclide release that is needed to assess both the dose and activity compliance levels with EPA's 40 CFR 190 regulations?

Iodine-129 may need to be included as a fission product in a source-term analysis; however, it is not one of the more important isotopes for accident analyses because of its long half-life and low specific activity. On the other hand, iodine-129 may be important for normal plant operations if it is routinely emitted from the reprocessing facility. The difference is that iodine-129 could build up in the environment over an extended period of time due to years of plant operation, depending on the fraction of iodine that is released routinely as opposed to being captured in the off-gas processing.

17.   We like that SNL is looking at different types of fuel, namely different cooling times and burn ups.

We will continue to take account of the fact that a range of fuel types and fuel ages are likely to be reprocessed.

18.   Need to check flow diagram on slide 6 of David Louie's presentation. Where was the data obtained and how was it developed?

To avoid making the presentation OUO, this information was not provided. It can be provided separately if needed, but the information would need to be treated as OUO.

19.   MELCOR would seem to be the best code for the task. However, we need to consider its limitations. For example, there is no plume rise model available. Is this a concern?

We agree that limitations of MELCOR and other codes need to be considered in the final evaluation and selection process, but that is part of our Task 3 that will be performed in the future.

Plume rise, in the context of an atmospheric release, is properly treated as part of a level-3 consequence analysis, so this is not an issue for a source-term tool. Plume rise, in the context of a fire model, is discussed under Comment 3.

20.   A definition of source term releases is provided on Slide 2 of Fred Gelbard's presentation. Should not this definition be expanded to include radioactivity levels (e.g., Curies) and gaseous releases (e.g., ^{85}Kr, ^{14}C).

We agree that gaseous releases should have been specifically called out. The source term tool needs to ultimately produce release information in terms of activities, and this should have been discussed as well.

21.   It is not clear if the aerosol model Fred Gelbard presented, actually shown as a nice movie, will be incorporated as part of the source term code. Will there be size limitations that need to be considered in running a model of this type from our desktops?

From the Sierra calculations, correlations for releases will be developed and either incorporated into the tool or a methodology for their inclusion in the analysis process will be provided to the NRC. The Sierra calculations typically require days of computation on a very large parallel computer, and hence are not appropriate for desktop computers.

2. TITLE AND SUBTITLE		3. DATE REPORT PUBLISHED	
Review of Spent Fuel Reprocessing and Associated Accident Phenomena		MONTH	YEAR
		February	2017
		4. FIN OR GRANT NUMBER	
		V-6050	

5. AUTHOR(S)	6. TYPE OF REPORT
N. E. Bixler, F. Gelbard, D. L. Y. Louie, and J. Phillips	Final
	7. PERIOD COVERED (Inclusive Dates)
	2015-2016

8. PERFORMING ORGANIZATION - NAME AND ADDRESS (If NRC, provide Division, Office or Region, U. S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)

Sandia National Laboratories
Albuquerque, New Mexico 87185

9. SPONSORING ORGANIZATION - NAME AND ADDRESS (If NRC, type "Same as above", if contractor, provide NRC Division, Office or Region, U. S. Nuclear Regulatory Commission, and mailing address.)
Division of Risk Analysis
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001

10. SUPPLEMENTARY NOTES

11. ABSTRACT (200 words or less)

The work presented in this report supports the advancement of the safety assessment capabilities of the NRC staff in terms of evaluating accidents that could release radioactivity into the environment and estimating the associated risk during the reprocessing of nuclear fuel. This report lays out much of the background and the concepts needed to construct a computational tool for reprocessing facility source terms. It reviews the following topics: (1) the status of past and current reprocessing plants throughout the world; (2) an overview of a typical plant design, its functions, and its layout; (3) historical accidents and the phenomena that are relevant to these accidents; and (4) the models that are needed to describe these accidents. This document does not propose or describe a specific computational tool that can be used to estimate source terms for a reprocessing facility. Rather, it supports the NRC's capability to evaluate potential accident source terms that would be used in a license application for a reprocessing facility.

12. KEY WORDS/DESCRIPTORS (List words or phrases that will assist researchers in locating the report.)	13. AVAILABILITY STATEMENT
Reprocessing, Nuclear Fuel, accident, spent nuclear fuel, accident phenomena, aerosol modeling	unlimited
	14. SECURITY CLASSIFICATION
	(This Page) unclassified
	(This Report) unclassified
	15. NUMBER OF PAGES
	16. PRICE

Printed
on recycled
paper

Federal Recycling Program

NUREG/CR-7232

Review of Spent Fuel Reprocessing and Associated Accident Phenomena

February 2017