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# TRITIUM FACILITY & EQUIPMENT DESIGN

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# NUC-133 EXAM PREVIEW

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## Exam Preview:

1. In Tritium gloveboxes it is important to minimize the in-leakage of air (oxygen) for flammability concerns (however no gloveboxes have been operated with negative differential since 1991, when the Tritium Task Group 28 recommended against this practice which was observed at the Salt Facility at Los Alamos).
  - a. True
  - b. False
2. Even at the relatively high leak rates of  $10^{-3}$  to  $10^{-4}$  cm<sup>3</sup> He/s, approximately 0.1 Ci of tritium will be released for every 1,000 to 10,000 seconds of elapsed time. This is a release of approximately 1 Ci from the facility stack for every 3 to \_\_\_ storage hours, which generally does not pose a significant health risk.
  - a. 10
  - b. 20
  - c. 30
  - d. 40
3. If the cleanup system is associated with a low-quality barrier (leak rate of \_\_\_ Ci or more per minute), the cleanup system flow rate needs to be very high so that tritium is removed from the gas before it is released to the environment.
  - a. 10
  - b. 4
  - c. 6
  - d. 8
4. In principle, nickel and nickel alloys is a suitable material in tritium systems.
  - a. True
  - b. False

5. All-metal mechanical joints are also a sound way to join components in tritium systems. Typically, copper, silver-plated nickel, or silver-plated stainless steel have been used as gaskets.
  - a. True
  - b. False
6. According to the reference material, in a second case, \_\_\_\_ failures out of six tests occurred when high-quality Type 316 stainless steel was exposed to tritium gas in the presence of Teflon TM shavings and 500 ppm moisture. All of the failures were catastrophic, and all were the result of massively induced stress-crack corrosion. The conditions that set up the introduction of the massively induced stress-crack corrosion in this case
  - a. 3
  - b. 4
  - c. 5
  - d. 6
7. The design requirements for tritium are a function of the tritium form, quantity, concentration, pressure, and period of storage. High concentrations of tritium gas stored at high pressure (> \_\_\_\_\_ psia) are difficult to contain due to tritium and helium embrittlement of the container materials.
  - a. 2,000
  - b. 3,000
  - c. 4,000
  - d. 5,000
8. Materials used in tritium should be fabricated in a manner that produces little contamination. Methods that are consistent with high vacuum processes are often useful. When joining is required, gas metal arc welding (GMAW) is the preferred welding method.
  - a. True
  - b. False
9. According to the reference material, temperatures above about \_\_\_\_°C accelerate radiation effects in polymers, so the temperature of any polymer parts should be kept as low as possible.
  - a. 90
  - b. 100
  - c. 120
  - d. 140
10. A tank at atmospheric pressure would have a final pressure of \_\_\_\_ pounds per square inch after all the tritium had decayed, so embrittlement is not an issue.
  - a. 15
  - b. 30
  - c. 45
  - d. 55

|                                                                                                  |        |
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## FOREWORD

Tritium handling practices have evolved over several decades at Department of Energy (DOE) tritium facilities. The objective has been to accomplish required tritium work while minimizing and controlling the exposure of workers, the public, and the environment to tritium. This document provides guidance for the handling, storing, and shipping of tritium.

This Standard is approved for use by all DOE elements and their contractors. DOE technical standards, such as this Standard, do not establish requirements. However, all or part of the provisions in a DOE standard can become requirements if either they are explicitly stated to be requirements in a DOE requirements document or the organization makes a commitment to meet the Standard in a contract or in an implementation or program plan. The DOE implementation plan to address the Defense Nuclear Facilities Safety Board (DNFSB) Recommendation 2005-1 committed to implement interim storage packaging provisions for tritiated materials, which are discussed in Section 6.6. These packaging provisions are requirements for tritium facilities under the auspices of the National Nuclear Security Administration (NNSA) and the Environmental Management Program Office (EM).

The author of the Standard, Bill Weaver of DOE Office of the Chief of Nuclear Safety (CNS), wishes to acknowledge the contributions of the Savannah River National Laboratory (SRNL) and the Savannah River Site (SRS) staff, Paul Blanton, Paul Korinko, Greg Staack, and Steve Xiao; Ken Keeler and Mike Rogers of the Los Alamos National Laboratory (LANL); Diane Spencer of the Lawrence Livermore National Laboratory (LLNL); CNS staff members Steve McDuffie, Marlene Fitzpatrick, and Elaine Beacom; Nazir Kherani of the University of Toronto; Armando Antoniazzi of Kinectrics, Inc.; Genevieve Weaver of Penn State; Tracy Getz, Robin Henderson and Robert Waxman of Office of The General Counsel; and Steve Zobel, a staff member from the DOE Office of the Associate Under Secretary for Environment, Health, Safety and Security.

## ACRONYMS

|                  |                                                                                      |
|------------------|--------------------------------------------------------------------------------------|
| AEA              | Atomic Energy Act of 1954                                                            |
| AI               | Alveolar-Interstitial region                                                         |
| ALARA            | As Low As Reasonably Achievable                                                      |
| ALI              | Annual Limit on Intake                                                               |
| AMAD             | Activity median aerodynamic diameter                                                 |
| AMD              | Activity median diameter                                                             |
| ASCE             | American Society of Civil Engineers                                                  |
| ASME             | American Society of Mechanical Engineers                                             |
| ASN              | French Nuclear Safety Authority                                                      |
| ANSI             | American National Standards Institute                                                |
| ARAR             | Applicable or Relevant and Appropriate (CERCLA)                                      |
| AU               | Office of the Associate Under Secretary for Environment, Health, Safety and Security |
| AWQC             | Ambient water quality criteria                                                       |
| bb               | bronchiolar region                                                                   |
| BB               | Bronchial region                                                                     |
| Bq               | Becquerel                                                                            |
| BS               | Bone surface                                                                         |
| BTSP             | Bulk Tritium Shipping Package                                                        |
| BZA              | Breathing zone air (sampler)                                                         |
| CANDU            | CANada Deuterium Uranium pressurized reactor                                         |
| CERCLA           | Comprehensive Environmental Response, Compensation, and Liability Act                |
| CFR              | Code of Federal Regulations                                                          |
| Ci               | Curie                                                                                |
| CMD              | Count median diameter                                                                |
| CoC              | Certificate of Compliance                                                            |
| CRC              | Combustion Research Center                                                           |
| CWA              | Clean Water Act                                                                      |
| DCS              | Derived Concentration StandardD&D    Decontamination and Decommissioning             |
| DAC              | Derived Air Concentration                                                            |
| DBA              | Design Basis Accident                                                                |
| DBE              | Design Basis Earthquake                                                              |
| DCF              | Dose Conversion Factor                                                               |
| DCF <sub>o</sub> | Dose Conversion Factor based on observed activity                                    |
| DCG              | Derived Concentration Guide                                                          |
| DNFSB            | Defense Nuclear Facilities Safety Board                                              |
| DOE              | U.S. Department of Energy                                                            |
| DOT              | U.S. Department of Transportation                                                    |

|                 |                                                     |
|-----------------|-----------------------------------------------------|
| DPM             | Disintegrations per minute                          |
| DSA             | Documented Safety Analysis                          |
| E <sub>50</sub> | Committed Effective Dose                            |
| EDL             | Economic Discard Limit                              |
| EPCRA           | Emergency Planning and Community Right-to-Know Act  |
| EPDM            | Ethylene Propylene Diene Monomer                    |
| EPA             | Environmental Protection Agency                     |
| EH              | Office of Environment, Safety and Health            |
| EIS             | Environmental Impact Statement                      |
| EM              | Office of Environmental Management                  |
| ET              | Extrathoracic                                       |
| f <sub>1</sub>  | fraction of radionuclide absorbed from the GI tract |
| FCA             | Fire Control Area                                   |
| FDTAS           | Field Deployable Tritium Analysis System            |
| FY              | Fiscal Year                                         |
| GI              | Gastrointestinal                                    |
| HEPA            | High-Efficiency Particulate Air                     |
| HDPE            | High-Density Polyethylene                           |
| HIVES           | Highly Invulnerable Encased Safe                    |
| HMR             | Hazardous Material Regulations                      |
| HSV             | Hydride Storage Vessel                              |
| HSWA            | Hazardous and Solid Waste Amendments                |
| HTV             | Hydride Transport Vessel                            |
| HVAC            | Heating, Ventilation, and Air Conditioning          |
| IAEA            | International Atomic Energy Agency                  |
| IATA            | International Air Transport Association             |
| ICRP            | International Commission on Radiological Protection |
| IMT             | Insoluble Metal Tritide                             |
| INL             | Idaho National Laboratory                           |
| ISM             | Integrated Safety Management                        |
| ITER            | International Thermonuclear Experimental Reactor    |
| ITP             | Insoluble Tritiated Particulate                     |
| keV             | Kiloelectron volt                                   |
| LANL            | Los Alamos National Laboratory                      |
| LDPE            | Low-Density Polyethylene                            |
| LDR             | Land Disposal Restriction                           |
| LLD             | Lower Limit of Detection                            |
| LLI             | Lower Large Intestine                               |
| LLNL            | Lawrence Livermore National Laboratory              |
| LN              | Lymph Node                                          |
| LSA             | Low Specific Activity                               |
| LSC             | Liquid Scintillation Counting                       |

|       |                                                    |
|-------|----------------------------------------------------|
| LLW   | Low-level waste                                    |
| MAR   | Material at Risk                                   |
| mCi   | Millicurie                                         |
| MCL   | Maximum Contaminant Level                          |
| mm    | Millimeter                                         |
| mrem  | Millirem                                           |
| NFPA  | National Fire Protection Association               |
| NMMSS | Nuclear Materials Management and Safeguards System |
| NNSS  | Nevada National Security Site                      |
| NP    | Nasal Passage Region                               |
| NPDWR | National Primary Drinking Water Regulation         |
| NPH   | Natural Phenomena Hazard                           |
| NRC   | U.S. Nuclear Regulatory Commission                 |
| NRPB  | National Radiological Protection Board             |
| OBT   | Organically Bound Tritium                          |
| OPI   | Office of Primary Interest                         |
| ORR   | Operational Readiness Review                       |
| OH-   | Hydroxide                                          |
| P     | Pulmonary Parenchyma Region                        |
| PC    | Performance Category                               |
| PCB   | Polychlorinated biphenyl                           |
| PMR   | Palladium Membrane Reactor                         |
| PPE   | Personal Protective Equipment                      |
| PPPL  | Princeton Plasma Physics Laboratory                |
| psia  | pounds per square inch absolute                    |
| psig  | pounds per square inch gauge                       |
| PSO   | Program Secretarial Officer                        |
| PTFE  | Polytetrafluoroethylene                            |
| PVC   | Polyvinyl chloride                                 |
| PV    | Product Vessel                                     |
| RCRA  | Resource Conservation and Recovery Act             |
| RCS   | Radiological Control Standard                      |
| RM    | Remainder Organ                                    |
| RMA   | Radioactive Materials Area                         |
| RTF   | Replacement Tritium Facility                       |
| RWP   | Radiological Work Permit                           |
| S     | Stomach or specific source organ (used with SEE)   |
| SAES  | Società Apparecchi Elettrici e Scientifici         |
| SAF   | Self Absorption Factor                             |
| SAM   | Surface Activity Monitor                           |
| SAR   | Safety Analysis Report                             |
| SCO   | Surface Contaminated Object                        |

|        |                                                                      |
|--------|----------------------------------------------------------------------|
| SDWA   | Safe Drinking Water Act                                              |
| SEE    | Specific Effective Energy                                            |
| SEL    | Seismic Equipment List                                               |
| SEM    | Scanning Electron Microscope                                         |
| SEP    | Seismic Evaluation Procedure                                         |
| SI     | Small Intestine                                                      |
| SMT    | Stable Metal Tritide                                                 |
| SNL    | Sandia National Laboratory                                           |
| SNLL   | Sandia National Laboratory, Livermore                                |
| SNM    | Special Nuclear Material                                             |
| SRNL   | Savannah River National Laboratory                                   |
| SRS    | Savannah River Site                                                  |
| SSCs   | Structures, Systems, and Components                                  |
| STC    | Special Tritium Compound                                             |
| Sv     | Sievert                                                              |
| T      | Tissue                                                               |
| TB     | Trachea and Bronchial Region                                         |
| TFG    | Tritium Focus Group                                                  |
| TRL    | Tritium Research Laboratory, Sandia National Laboratory              |
| TWD    | Technical Work Document                                              |
| TSD    | Treatment, Storage, and Disposal                                     |
| TSR    | Technical Safety Requirement                                         |
| UB     | Urinary Bladder                                                      |
| UHMWPE | Ultra-High-Molecular-Weight Polyethylene                             |
| ULI    | Upper Large Intestine                                                |
| WETF   | Weapons Engineering Tritium Facility, Los Alamos National Laboratory |
| WSRC   | Washington Savannah River Company                                    |

## 4.0 FACILITY DESIGN

DOE tritium facilities, as a subset of DOE nonreactor nuclear facilities, must conform to the design requirements contained in DOE O 420.1C, *Facility Safety*. Implementation guidance for this Order can be found in DOE G 420.1-1A. The general design philosophy of nuclear facilities in general, and tritium facilities specifically, follows the tenets of multiple levels of protection; preventive vs. mitigative design features, passive vs. active safety systems, engineered vs. administrative controls, and judicious use of zoning throughout the facility.

### 4.1 Confinement of Tritium Philosophy

It is desirable to have tritium construction projects managed by a team that includes tritium operations expertise on staff. The building/systems would be designed to meet the needs of the user, and costly retrofits after completion could be avoided.

During the first 25 years of tritium technology, the handling techniques in use were designed to protect the worker from exposure to tritium. Worker protection was provided primarily by the use of single-pass ventilation systems designed to rapidly remove any tritium released in the breathing space from the area occupied by the workers. The ventilation gases were released through an elevated stack at high velocity to massively dilute the gases before they could reach ground level. Single-pass ventilation systems and high-velocity hoods were used extensively and quite successfully for worker protection during these early years. These high-velocity ventilation, high-velocity air hood, and elevated release techniques are still used for worker protection, but generally as a supplement to improved barriers that better protect the environment.

In those early years, the room or building enclosing the tritium activity was equipped with a single-pass ventilation system that did not recirculate the air back into the facility. Outside air was brought in by the ventilation fans, conditioned for comfort, passed through the building spaces one time, and was then released to the environment through an elevated stack. The room air exchange rate generally accepted to be adequate for worker protection was 6 to 10 room air changes per hour.

The tritium apparatus was enclosed in a high-velocity air hood, and the worker worked through gloves in the doors or reached in through hood openings to operate the equipment. The high-velocity air hoods were maintained at a pressure negative to the room spaces, and the natural air

flow was from the room through the hood opening and then out the ventilation duct work and up the stack to the environment.

Tritium releases that occurred due to normal operations, component failure, and worker error were inside the hood. The high-velocity air flowing through the hood swept any released tritium away from the worker and out the stack to the environment where the tritium was massively diluted in concentration. These techniques protected the worker; however, tritium was released to the environment.

As concern for the environment increased, tritium technologists first attempted to control releases by increasing design and material selection requirements, adding and enforcing regulations, and increasing worker training and awareness. Time would prove that these techniques, although helpful, were not completely successful. The tritium workers were already operating at a high performance level and the tritium releases associated with equipment design and material selection were not measurably decreasing as a result of the more stringent design requirements and reviews. Tritium releases continued to occur. The expectation of faultless materials and errorless workers was unrealistic. By the late 1960s and early 1970s, another philosophy, one of capture, containment, and cleanup evolved.

#### 4.1.1 Tritium Capture, Contain, and Cleanup Process

The capture, contain, and cleanup process encloses the primary or first wall tritium container inside a secondary container such as a double-walled container, glovebox, room, or building so that any tritium escaping from the primary container is captured in the secondary container. A tritium removal system associated with the secondary container then removes the tritium from the secondary by circulating the captured gases through a cleanup system.

##### 4.1.1.a Containment and Confinement Systems

There are many different technical descriptions of the terms containment and confinement. The simple dictionary definitions are: containment – being contained, which in turn is to hold or enclose; and confinement – being confined, which in turn is to restrict, to keep within limits. It is beneficial to define how these words are used in the tritium field. In some facilities and applications, the terms are used interchangeably. The TFG definitions are defined in Appendix B, *Definitions*. This Standard as noted in the Definitions makes a distinction between these terms as follows:

**Containment system:** A collection of passive barriers that can satisfy a specified leak criterion without operation of any ancillary equipment. An example of a containment system is a series of piping and vessels enclosing tritium gas operations. An example of a simple double containment system is a container within another container with each container acting as a separate and

independent containment system; more intricate double containment systems have the capability to monitor the volume between the containers for leak detection of the inner container.

**Confinement system:** A collection of barriers that can satisfy a specified leak criterion contingent upon operation of its ancillary (active) system. Examples of confinement systems include: a glovebox and its associated cleanup system, and a room with its associated cleanup system.

Simply stated, containment consists of some arrangement of physical barriers that do not need any other equipment or operator action to operate to meet a leak rate criterion, whereas confinement consists of an arrangement of barriers (any kind) that need an active system or action to meet its leak rate criterion. Note that in the context of these definitions, a glovebox with an associated glovebox cleanup system is a confinement system. A glovebox structure itself is a containment system if, and only if, the specified leak criterion can be met by the structure itself.

As mentioned, the terms containment and confinement are not well defined in the DOE complex. For example, DOE M 435.1-1 defines confinement as follows:

Confinement: The control or retention of radioactive materials within a designated boundary. Primary confinements are process enclosures and other spaces normally containing radioactive material. Secondary confinement surrounds one or more primary confinement systems.

Adding to the confusion are requirements for ventilation systems. DOE G 420.1-1A for use with DOE O 420.1C does not define containment or confinement, but defines the terms primary, secondary, and tertiary “confinement barriers” as:

- Primary confinement. Provides confinement of hazardous material to the vicinity of its processing. This confinement is typically provided by piping, tanks, gloveboxes, encapsulating material, and the like, along with any off-gas systems that control effluent from within the primary confinement.
- Secondary confinement. Consists of a cell or enclosure surrounding the process material or equipment along with any associated ventilation exhaust systems from the enclosed area.
- Tertiary confinement. Typically provided by walls, floor, roof, and associated ventilation exhaust systems of the facility. It provides a final barrier against the release of hazardous materials into the environment.

The Guide also references DNFSB/TECH 34, *Confinement of Radioactive Materials at Defense Nuclear Facilities*, in which the terms “active confinement” systems and “passive confinement”

systems are discussed. The terms “active confinement” and “passive confinement” roughly translate to confinement and containment, respectively, as used in this Standard.

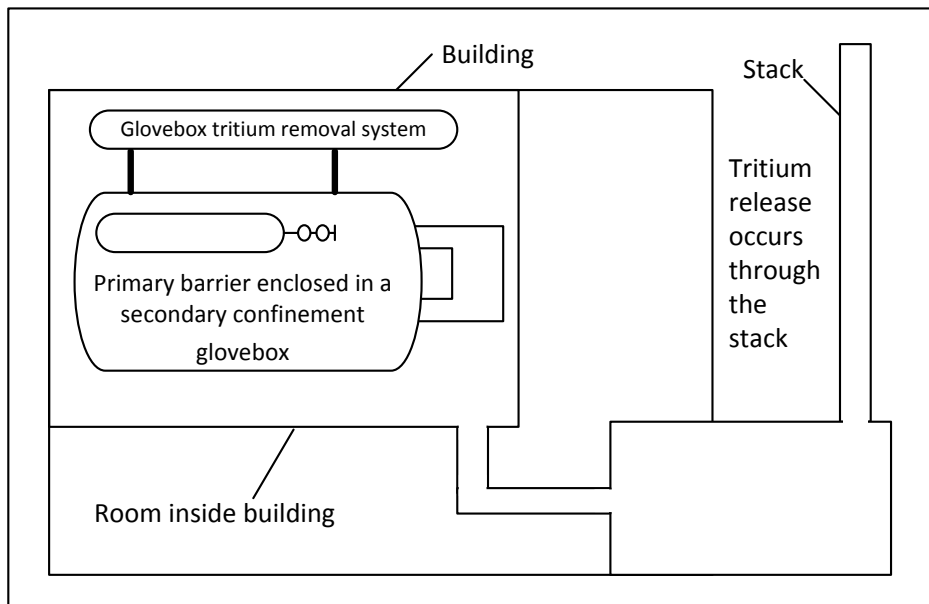
#### 4.1.1.a(1) Primary Containers

Operations are conducted with tritium enclosed inside leak tight primary containers consisting of parts such as valves, tubing, pipe, components, transducers, pumps, and vessels. The leak rate of these primary or first wall containers, at operating pressure, is generally certified to be less than  $10^{-6}$  to  $10^{-7}$  cm<sup>3</sup> of helium per second (cm<sup>3</sup> He/sec). The quantity of tritium released during all normal operations is extremely small and can be estimated from the engineering specifications. Many primary tritium systems are designed with pressure relief protection. These devices should not relieve directly into the environment, but rather into holding tanks designed with sufficient capacity to retain the entire contents of the primary system.

#### 4.1.1.a(2) Secondary Containers

In modern tritium operations, the primary container is enclosed inside a secondary barrier such as a glovebox. The secondary system is only exposed to tritium, if it is released from the primary barrier.

Secondary containers in the DOE complex vary in size, shape, leak rate, and quality depending upon the age, projected use, and the quantity of tritium at risk. Figure 4-1 illustrates an example of secondary containment.



7) Figure 4-1: Secondary containment enclosing a primary container filled with tritium inside a building equipped with single-pass ventilation to a stack

#### 4.1.1.a(2)(a) High-Quality Secondary Containers

Some operations are equipped with high-quality, double-walled pressure vessels. The outer pressure vessel is the secondary containment system, and the inner container is the primary container. This type of secondary containment system is generally used for storage of large quantities of tritium of very high quality, and is certified at operating pressure to leak rates of less than  $10^{-6}$  to  $10^{-7}$  cm<sup>3</sup> He/s.

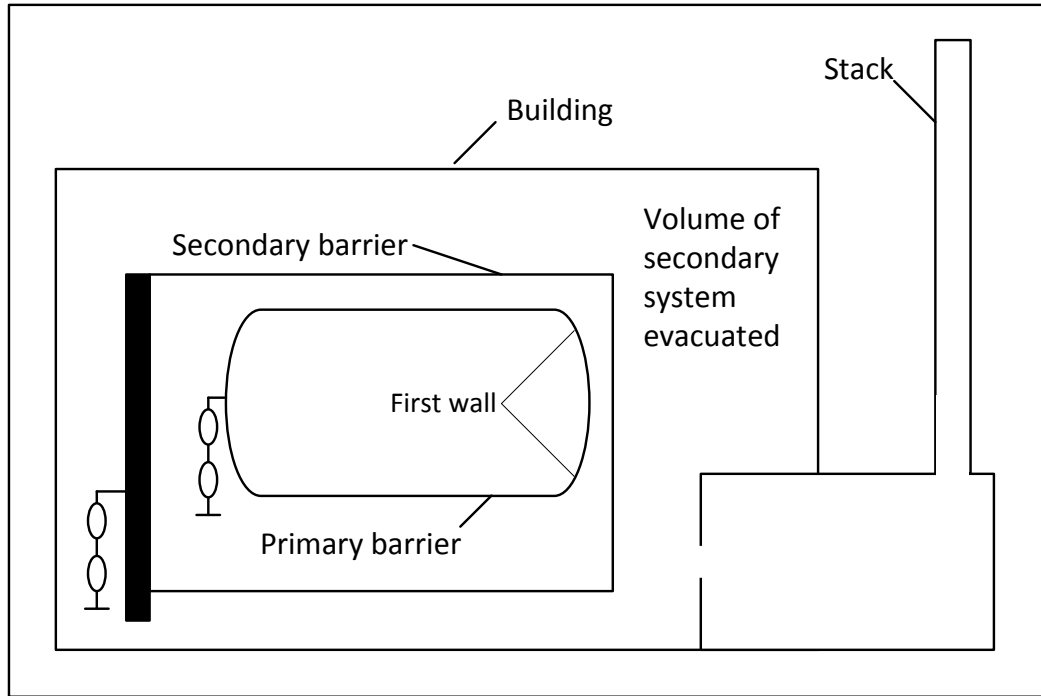
These high-quality secondary containment systems safely store any tritium released from the primary container for several days or weeks without a significant release to its environment. The maximum quantity of tritium released from these high-quality systems during a significant primary container leak can be accurately estimated from the secondary containment system engineering specifications.

The space between the primary vessel and the secondary container in these systems is usually evacuated during service. If tritium is released into these spaces, there are no dilution gases present, and the gas leaking from the secondary container is in the same form as the gas in the primary container. If the gas released into this high-quality secondary is 90 percent tritium, less than 1 Ci of tritium will be released to the surrounding area for each four to 40 storage days.

Following a release into a high-quality secondary container, the tritium can be recovered by pumping it from the secondary container into another primary container. Several days can elapse during the recovery process without a significant release of tritium to the environment.

#### 4.1.1.a(2)(b) Medium-Quality Secondary Container

It is not practical or possible to enclose all primary tritium containers inside high-quality, non-diluting, evacuated secondary containers. Gloveboxes, as discussed in 4.1.1.a, can be utilized in both secondary containment and secondary confinement systems. Figure 4-2 illustrates one example.



8) Figure 4-2: Secondary confinement

Most primary containers can be enclosed in gloveboxes. The box gives access to the primary containers for ease of operation and maintenance.

The leak rate of a glovebox can generally be certified to be no more than  $10^{-3}$  to  $10^{-4}$   $\text{cm}^3$  He/s. In a typical working environment, tritium gloveboxes are operated at near room pressure but slightly negative, on the order of a few tenths of an inch of water column. Tritium gloveboxes do not operate at the large differential pressures common with other gloveboxes through the DOE complex such as those that house actinides and fission products. In Tritium gloveboxes it is important to minimize the in-leakage of air (oxygen) for flammability concerns (however no gloveboxes have been operated with positive differential since 1991, when the Tritium Task Group<sup>28</sup> recommended against this practice which was observed at the Salt Facility at Los Alamos). The low differential pressure allows for less potential for in-leakage than for the larger differentials gloveboxes but the oxygen monitors in the tritium gloveboxes are usually still credited in the safety basis. Although the glovebox is still negative with respect to the room, the tritium levels in the box are usually several orders of magnitude greater than the tritium levels in the room and, due to the laws of partial pressures, the movement of tritium due to permeation alone will always be from the box to the room. To minimize the permeation of tritium from the box to the room, gloveports should be covered and evacuated when they are not in use.

<sup>28</sup> U.S Department of Energy, *Report of the Task Group on Operation of Department of Energy Tritium Facilities*. October 1991.

In addition to oxygen monitors, most tritium gloveboxes are connected to cleanup systems designed to remove undesired impurities from the glovebox gases and return clean gases back to the box. One of the undesired impurities is tritium (as T<sub>2</sub>, HT, and/or HTO) itself. Although cleanup systems are designed to remove free tritium down to the part-per-million to part-per-billion level the return gases from such cleanup systems will never be completely devoid of free tritium. In addition, cleanup systems designed to remove free tritium from the glovebox gases are not capable of removing tritium that has plated-out on the interior surfaces of the glovebox or any of the equipment that is inside the glovebox. Thus, when the box is opened to room air for maintenance purposes, a “puff” type of tritium release will occur, such as that described in Section 2.3.5 “Outgassing”. The chemical form of the tritium release will be HTO.

Tritium permeation and diffusion through the elastomeric seals and gloves is reasonably well known, and the tritium released from the glovebox during a primary container leak can be accurately estimated.

If tritium is released into the glovebox, the gases in the glovebox are mixed with and dilute the tritium. The gases leaking from the glovebox are then in the form of tritium mixed with the glovebox gases. Assuming a glovebox with a volume of 1 m<sup>3</sup> and a release of 10 grams of tritium, the glovebox concentration following the release will be 0.1 Ci/cm<sup>3</sup>. Even at the relatively high leak rates of 10<sup>-3</sup> to 10<sup>-4</sup> cm<sup>3</sup> He/s, approximately 0.1 Ci of tritium will be released for every 1,000 to 10,000 seconds of elapsed time. This is a release of approximately 1 Ci from the facility stack for every 3 to 30 storage hours, which generally does not pose a significant health risk.

Following a release into a glovebox, the tritium is recovered in a tritium removal system. This low-level waste is in the form of water contaminated with tritium. Several hours can elapse during the recovery process without a significant release of tritium to the environment.

#### 4.1.1.a(2)(c) Low-Quality Secondary Containers

Other facilities are equipped with lower-quality systems such as rooms or buildings. It is difficult to determine the leak rate of these low-quality systems and, therefore, it is difficult to estimate the quantity of tritium that will be released to the environment during a primary container leak.

If the secondary container is a room or building, the ventilation system could be shut off to reduce the release of the tritium to the environment through the building stack when a release occurs. In this case the released tritium contaminates the building. If a tritium cleanup system is available (in a room or a building), the ventilation system is switched over to the recirculation mode to reduce the amount of contamination. If a cleanup system is not available then a choice is made (ahead of time so it is proceduralized and personnel are trained) whether to stack or contain the release.

For example, if there is no cleanup system and the ventilation system is shut off, assuming a 100 m<sup>3</sup> (20 × 18 × 10 feet) building and a 1-gram release to the building, the tritium concentration in the

building gases will be  $0.0001 \text{ Ci/cm}^3$ . If the building is the containment barrier, it will leak approximately 5 percent of its volume to the environment per hour; therefore, an environmental tritium release of 500 Ci/hr will result. Cleanup of the building also is required in this scenario.

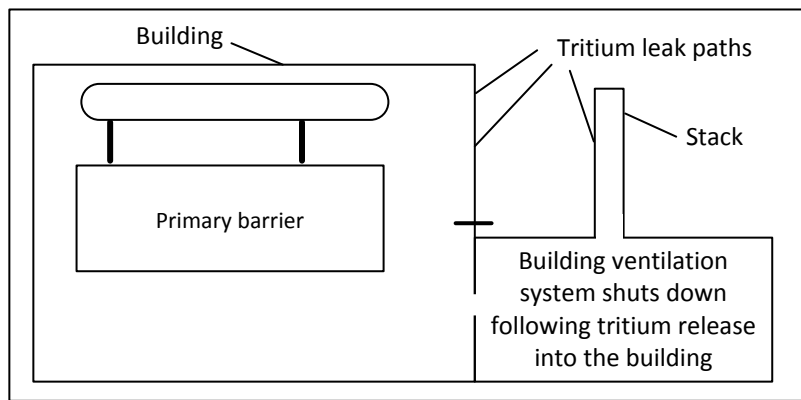
Even with a room or building cleanup system, a substantial fraction of the tritium released from the primary containment system will be released to the environment before recovery is accomplished due to the high leak rate of these low quality secondary systems. However, these low-quality systems still have a place in the tritium containment strategy. These room- or building-type systems, equipped with high flow rate tritium removal systems, are used in facilities where there are no other feasible alternatives due to the large size of the equipment being enclosed.

#### 4.1.1.a(3) Primary and Secondary Containers

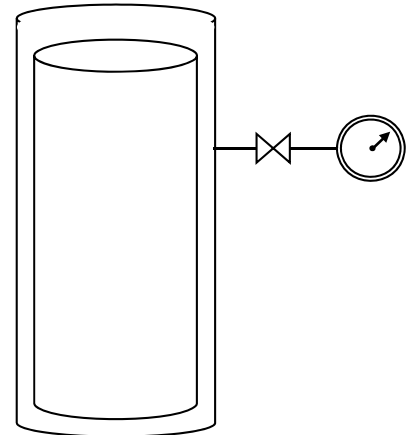
The container-within-a-container concept introduced above is applied in the tritium community in various configurations. Figure 4-3.a shows a building confinement system. Figure 4-3.b shows an arrangement consisting of a higher-quality primary container with a lower-quality secondary container with indication or sampling capability from the annulus area to help detect whether the primary is leaking. Before opening the secondary, the annulus area should be sampled and if a leaking primary is suspected from the sample or from the pressure indicator, then the container is moved to within a glovebox or compensatory measures taken prior to opening. Depending on the application and quality of the primary container, the secondary may not need to be a containment barrier itself. The secondary's for high quality primary containers are most often employed as a physical protection barrier for the primary container. For example, the LP-50 SARP<sup>29</sup> states...“no credit is taken for the seal on the shell in the safety analysis and no test of its seal tightness is made. The secondary vessel is a mechanical protection barrier. For primary containers of questionable or low quality, the secondary should provide a containment function as the probability of primary container failure is higher.” Section 6.6 discusses the container requirements for interim storage of tritium/ tritiated materials.

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<sup>29</sup> LP-50 SARP, *Safety Analysis Report for Packaging (SARP): USA/9507/BFL (ERDA-AL), Model AL-M1, MLM-2447*, September 30, 1977.



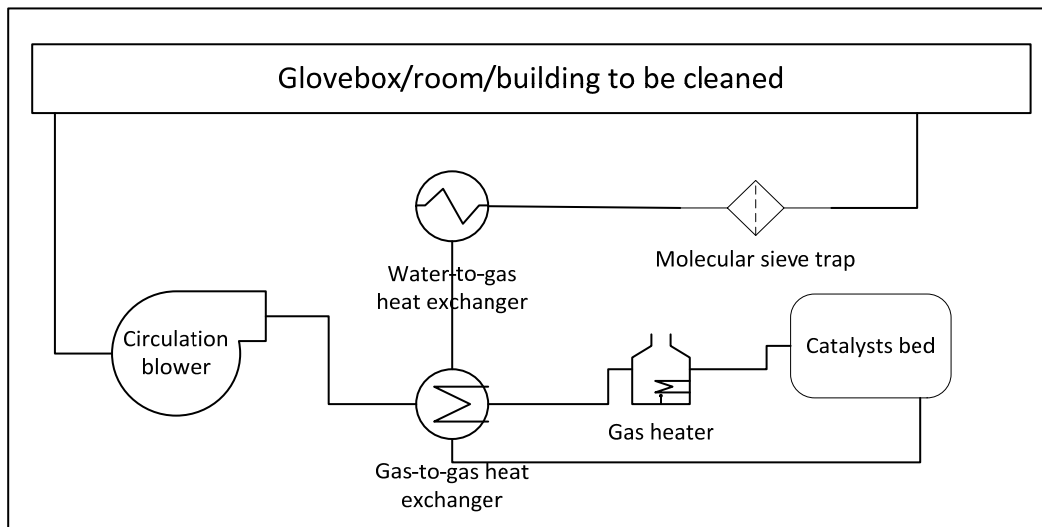
10) Figure 4-3.a: Building confinement system



9) Figure 4-3.b: Typical double containment configuration

#### 4.1.2 Tritium Cleanup and Removal Systems

Current designs employ tritium removal systems. When tritium is released into a secondary, the associated cleanup system starts, and the gases containing tritium are circulated through the cleanup system, and the tritium is removed. See Figure 4-4 for a diagram of a typical gas-to-water conversion tritium removal system.



11) Figure 4-4: Typical gas-to-water tritium removal system flow schematic

If the cleanup system is associated with a high-quality barrier (leak rate of less than 1 Ci of tritium in a period of 4 to 40 days), tritium transfers to another container can take several days to complete without a significant release of tritium to the environment. If the cleanup system is associated with a medium quality barrier (leak rate of less than 1 Ci in 3 to 30-hours), the cleanup system flow rate needs to be high enough to remove tritium within a few hours in order to prevent a significant release of tritium to the environment. If the cleanup system is associated with a low-quality barrier (leak rate of 8 Ci or more per minute), the cleanup system flow rate needs to be very high so that tritium is removed from the gas before it is released to the environment.

The captured tritium is generally removed by circulating the gas through a system that removes tritium down to the part-per-billion level. Typical present-day cleanup systems remove tritium by cracking the molecules on a hot catalyst. The free hydrogen atoms combine in the catalytic reactor with the oxygen present to form water. The tritiated water is then removed from the gas by molecular sieve traps. One example of this type of cleanup system that has been used extensively in several different tritium facilities works as follows:

- (1) Tritium is released into the secondary containment system (i.e., the glovebox) as a result of a primary system failure
- (2) The cleanup system is started and the tritium-containing gases captured in the glovebox are circulated through the cleanup system
- (3) The cleanup system removes the tritium from the gas stream by breaking down the hydrogen (i.e., H, D, and T) containing molecules on a hot, precious metal catalyst
- (4) The free H, D, and T atoms are recombined with oxygen in the catalytic reactor to form water vapor
- (5) The hot water vapor is cooled to a suitable temperature by passing the gas stream through one or more heat exchangers
- (6) The water is removed from the gas stream by passing the gas stream through molecular sieve traps

Catalyst/molecular sieve tritium removal systems of this type can be very effective. Depending on the tritiated species, the reduction in tritium concentration for such systems has been measured at ratios of  $10^6:1$  to  $10^8:1$  when operated in a once-through flow mode, in which the gas stream passes from the glovebox, through the cleanup system, and out the stack to the environment. In most situations, these types of cleanup systems are operated in the continuous flow mode, where the gas stream is moved from the glovebox, through the cleanup system, and back to the glovebox. When operated in the continuous flow mode, these types of cleanup systems can easily reduce the tritium concentration in the gas stream to the parts-per-billion level (i.e.,  $2.6 \text{ mCi/m}^3$ ), or lower, as long as they are operated for a sufficiently long period of time.

The tritium released to the environment during this process is a function of the quantity of tritium released, the volume and leak rate of the container, and the cleanup rate of the tritium removal system.

There are several considerations in determining the adequacy of the tritium removal system. First is the volume: the larger the volume, the longer it will take to remove tritium from the gases. Second is the tritium removal system flow rate. This rate affects the time required to remove tritium from the gases. The lower the flow rate, the longer it will take to remove tritium from the volume. Third is the cleanup rate of the tritium removal system. This system is typically operated in a recirculating mode, but may also be operated in a single-pass mode. The tritium-contaminated gases are pumped through the tritium removal system, and the cleaned gases are either returned to the volume or released to the environment.

In a large, complex system filled with equipment, it is difficult to know exactly how the returned or make-up gases will mix with gas in the system. The gas exit port should be spaced several feet away from the return port. It could be assumed that gas flows through in a slug, like a piston, and that only a single pass would be required to remove all of the tritium from the gases. Slug or piston displacement flow, however, is unlikely, and a more common assumption is to assume that the incoming tritium free gases exponentially dilute the gases.

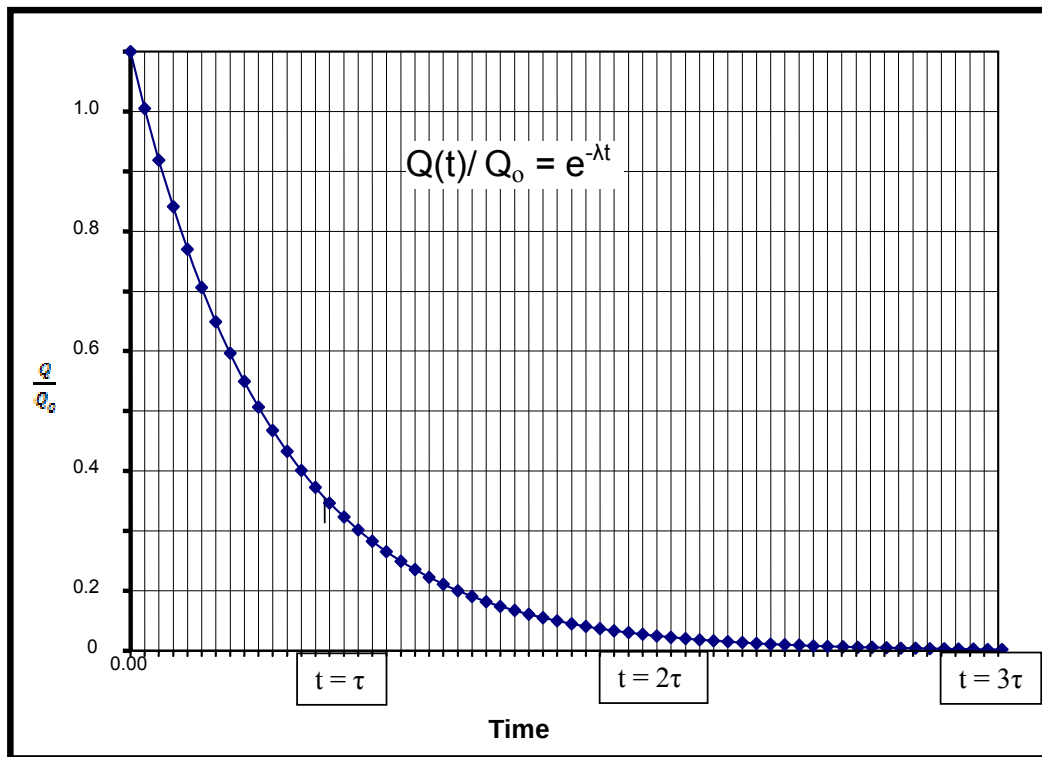
If uniform mixing is assumed, the resulting dilution at any time,  $t$ , from an initial quantity ( $Q_0$ ) of tritium due to operation of an air cleanup system can be estimated with standard exponential methodology. The return air from the cleanup system is also assumed to be completely detritiated.

$$Q(t) = Q_o e^{-\lambda t}$$

Where  $Q_o = Q(t = 0)$

where  $\lambda$  is the decay constant associated with the ventilation system equal to  $F/V$ , where  $F$  is the flowrate of the tritium removal system and  $V$  is the volume of the room cleaned. The graph of an exponential decay process is illustrated in Figure 4.5.

The quantity of tritium vs time is depicted in terms of time constants  $\tau$  or  $1/\lambda$ . When  $t =$  one time constant, roughly 63% of the tritium in the room has been removed; at 3 time constant, 95% removal has been achieved.



12) Figure 4-5: graph of an exponential decay process

$\lambda$  is a combination of three terms:  $\lambda_R$  (removal system)  $\lambda_L$  (leakage) to account for leakage from the room to the environment and  $\lambda_D$  (radioactive decay) of tritium in the room, yielding

$$Q_t = Q_o e^{-t(\lambda_R + \lambda_L + \lambda_D)}$$

Since tritium's radioactive half-life is 12.3 years,  $\lambda_D$  is normally ignored; however, as discussed in section 2.3.5, Outgassing, there could be some situations where outgassing can occur over a number of months or even years (the Pantex release of 1989 discussed in Section 4.7.7 is an example of this situation), in which  $\lambda_D$  can affect the calculated results; however, since ignoring the term is conservative, there is little need for further consideration here; yielding

$$Q_t = Q_o e^{-t(\lambda_R + \lambda_L)}$$

Note that the terms  $\lambda_R$  and  $\lambda_L$  both reduce the quantity of tritium in the room. In order to reduce the amount of tritium leaking to the environment for any given room configuration with an associated leak path factor, the tritium removal system flowrate (F) must be increased. Design of room and cleanup systems is an iterative process with a higher flow rate cleanup system compensating for a leaky room and vice-versa.

#### 4.1.3 Future Directions in Tritium Removal and Cleanup

Future tritium facilities should analyze the applicability of confinement systems in their facilities. For example, DOE O 420.1C states,

Confinement design must include the following:

- a. For a specific nuclear facility, the number, arrangement, and characteristics of confinement barriers as determined on a case-by case basis.
- b. The type, quantity, form, and conditions for dispersing the radioactive material in the confinement system design.
- c. An active confinement ventilation system as the preferred design approach for nuclear facilities with potential for radiological release [footnote omitted]. Alternate confinement approaches may be acceptable if a technical evaluation demonstrates that the alternate confinement approach results in very high assurance of the confinement of radioactive materials.

The guidance for confinement ventilation systems and evaluation of the alternatives, is provided in DOE Guide (G) 420.1-1A, *Nonreactor Nuclear Safety Design Guide for Use with DOE O 420.1C*, Facility Safety.

As regulatory release criteria and ALARA concerns are strengthened, the desirability of barriers increases. Many past confinement systems were of the glovebox and room or building design. One disadvantage of these systems is that tritium is converted to oxide, which must eventually be handled and disposed (with the attendant risks). Room- or building-type confinement systems were used in some tritium facilities such as in the T-building at Mound, in TFTR at Princeton Plasma Physics Laboratory (PPPL), and in TSTA and Weapons Engineering Tritium Facility (WETF) at Los Alamos. If the system is not very close to 100 percent efficient, the released oxide (which is about 10,000 times more toxic than the gas) could give an overall detriment (e.g., if 10,000 Ci of gas are collected, but only one Ci of oxide is released by the system, the whole operation is just a draw). Some current designs, notably the cleanup systems at the SRS H-Area New Manufacturing Facility, make use of gettering without oxidation. The primary advantage of these getter systems is that tritium is removed and stored in elemental form and is not converted by the tritium removal system to the more radiotoxic tritiated water. The application for glovebox atmospheres is good; however, severe challenges for removing tritium from room atmospheres with the required flow rates, while not fouling the getters, appear less promising. Research into other than metal (e.g., fullerene) gettering material has been sponsored by DOE in the past; subsequently, fullerenes have been

examined for hydrogen storage for the Hydrogen Economy<sup>30</sup>. Results indicate that stored tritium is stable for almost 10 years. The Loutfy et al. reference in section 1.4 presents an excellent overview on fullerenes.

It is possible that advances in design will make room or building-type confinement systems desirable, or it may turn out that these cleanup systems in general are found not to be cost-effective, and that a better use of resources to decrease environmental releases could be made by upgrading the existing primary and secondary systems. Additionally, there may be future glovebox decisions in which the oxidation process is still chosen over the gettering process due to programmatic reasons. There is no compelling agreement at this time to use room or building cleanup systems.

#### 4.1.4 Inspection and Surveillance Requirements

Instrumentation should be provided to monitor the leak-tight integrity of process piping, tanks, and other equipment. The surveillance measurements may include those of pressure differentials or flow rates that relate to the design leak rate. Radiation monitoring instrumentation may be used to qualitatively assess changes in leak rate.

### 4.2 Separation and Purification of Tritium

Isotopes have very similar chemical properties; therefore, their separation is, in general, more difficult than chemical separation. Thermal diffusion, fractional absorption, and cryogenic distillation have been used to separate tritium from protium and deuterium. Cryogenic distillation is the most proven separation technology; it is currently used in various locations including the Darlington Tritium Removal System and is the baseline for ITER. Cryogenic distillation is truly continuous and easily scalable. Gaseous thermal diffusion, especially for smaller scale operations, has been used successfully in several countries. Other separation techniques are also in use. Materials such as palladium were known having hydrogen isotopic effects shortly after the discovery of deuterium in 1931 and tritium in 1934. Batch palladium chromatography for hydrogen isotope separation dates to the 1950s. However, a “continuous” process using Pd chromatographic separation did not appear for half a century until Dr. Myung W. Lee of Savannah River National Laboratory (SRNL, then called SRL) originated the breakthrough concept of a “continuous” chromatographic separation process in 1979. The process was successfully developed into a working unit in the 1980s and subsequently named the Thermal Cycling Absorption Process (TCAP). Because TCAP demonstrated unequivocal advantages, it has gradually replaced all other separation processes for hydrogen isotope production and is currently the sole process of purifying tritium at SRS.

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<sup>30</sup> R.O. Loutfy, E. M. Wexler. *Feasibility of Fullerene Hydride as a High Capacity Hydrogen Storage Material*, Proceedings of the 2001 DOE Hydrogen Program Review, DOE 2001.

TCAP is a gas chromatograph in principle using palladium in the column packing, but it is unique in the fact that the carrier gas—hydrogen—is being isotopically separated and the system is operated in a semi-continuous manner. The aspects of hydride technology that provide interim hydrogen storage, generate pressure, and create a vacuum have been innovatively applied to create the TCAP process. During operation, thermal cycling moves gas back and forth in the Pd/k column, achieving efficient isotope separation at each of the column's separation stages.

During each cycle, a gas mixture is fed into the Pd/k column, and pure product and raffinate are withdrawn from the two ends of the column. The TCAP invention is an engineering achievement with simple design and advanced control logic and is suitable for radioactive confinement. A simple on/off valve is the only mechanical moving part. The robust design presents features of inherent safety and extra-long life. It can be scaled from very small to large with versatility depending on throughput. Testing has demonstrated that TCAP can separate the naturally occurring 150 ppm D<sub>2</sub> in a standard hydrogen cylinder, thus producing pure protium.

#### **4.3 Building Ventilation System**

If tritium does penetrate its barriers, it can be released into the worker breathing space. In this scenario, the ventilation system should be designed to meet the following objectives:

- Move released tritium from the worker breathing space as soon as possible;
- Minimize the contamination of other areas while moving released tritium;
- Release the tritium-contaminated gases at an elevation and velocity that will result in massive dilution and mixing with outside air before the tritium reaches ground level.

The ventilation system may be designed using the following guidelines:

- The ventilation system can be a single-pass ventilation system. Outside air is brought in through a supply fan, conditioned for the comfort of the workers, passes through the ductwork to the ventilated spaces one time, goes through the exhaust ductwork to an exhaust fan, and is released to the environment through the facility stack. Room recirculation designs can also be used in conjunction with single-pass ventilation systems, the design and desirability of which is discussed in Sections 4.1.2 and 4.1.3;
- The air supply and exhaust systems should be designed to eliminate dead air spaces where tritium-contaminated gases may accumulate;
- The ventilation system ductwork should not be shared with non-tritium operations;

- To minimize cross-contamination from one room to another, the exhaust gas from each room should dump into a central exhaust duct. Exhaust gases from several rooms should not be combined before being dumped into the central exhaust duct;
- To minimize cross-contamination from one ventilation function to another, the gases for each type of function, such as room ventilation, high velocity air hood ventilation, and glovebox ventilation, from a single room should not be combined until they reach the central exhaust duct;
- To ensure good mixing and dilution by outside air, the exhaust gases should be released to the environment through an elevated stack at high velocity;
- The ventilation system should be designed to use pressure zone control to minimize cross-contamination. The ventilation control system is designed to hold the spaces occupied by the tritium operations at a negative pressure relative to the spaces surrounding the facility. If the whole building is a tritium facility, then the building is at a slightly negative pressure relative to the environment. If the tritium is in a single room in a building, the room is at a negative pressure relative to the adjacent rooms. Air continuously leaks into the tritium operating areas from the surrounding environment. The ventilation zones of tritium processing areas should be maintained under controlled temperature and humidity conditions at all times to reduce the in-leakage of moisture to inert atmosphere gloveboxes. (Further reductions of in-leakage of moisture into inert gloveboxes can be achieved by selecting a glove material that has a low permeation rate for moisture and by reducing the exposure time (e.g., glove port covers) and/or total glove surface area);
- The walls separating adjacent rooms should be reasonably sealed to minimize tritium released from contaminating an adjacent room. Administrative controls need to be in place to require caulking and sealing around wall penetrations such as conduit and piping;
- For the ventilation system to work as designed, the inside and outside doors and airlocks need to be used properly. Propping an outside or inside door open will upset the pressure zone control system. An outside door bypasses the stack and provides a path for a ground level tritium release. The doors should be equipped with automatic door closures, and all personnel should be instructed on the proper use of doors;
- For facilities handling gram quantities of tritium, a rule of thumb is 6 to 10 air changes per hour as the standard of performance;
- The ventilation rate should be based on analysis of the hazards of the operations. A facility that has the potential to release only a few curies of gaseous tritium into the breathing space does

not need to operate at the same ventilation rate as a facility that has the potential to release larger quantities of tritium into the breathing space;

- Single-pass ventilation systems are expensive to operate because all air must be conditioned to make its single pass through the facility, and this conditioned air is not reused. Additionally, a high flow rate is desired in order to remove any released tritium from the facility as soon as possible to protect the workers;
- Special precautions such as personal protective equipment (PPE)—including respiratory protection and passing exhaust through particulate filters—may be needed when working with SMTs. D&D work with SMTs should be confined to occur within the workspace, if possible.

It is feasible to design future ventilation systems to operate at a variable flow rate that is a function of the time of day and the measured tritium concentration in the rooms. This would entail a higher initial cost, but would decrease the long-term operating costs without significant impact on facility safety. Additionally, the ventilation rate should be based on analysis of the hazards of the operations. For example, a facility that has the potential to release only a few curies of elemental tritium into the breathing space does not need to operate at the same ventilation rate as a facility that has the potential to release larger quantities of elemental or tritium oxide into the breathing space.

#### **4.4 Chilled Water System**

The chilled water system that is used to cool the tritium-related activities in a facility should be carefully designed to minimize the volume and tritium concentration of the contaminated water generated. The use of single-wall, water-to-gas heat exchangers in tritium removal systems and vacuum furnaces for example, will result in tritium contamination of the chilled water system. In some facilities, the same chilled water system is used to cool non-tritium activities in the same building, and, at some sites, the same chilled water system is also used in non-tritium related activities in adjacent buildings. As a result, tritium-contaminated wastewater is generated, and tritium contamination is spread from one piece of equipment to another, from one room to another, and from one building to another through the chilled water supply. This can lead to loss of control of a radioactive material.

The chilled water system should be designed to minimize the volume of water that can be contaminated with tritium. One technique is to use water-to-water heat exchangers or double-walled, gas-flushed water-to-gas heat exchangers to isolate the high volume central system from the tritium-related activities. The volume of water in the secondary loop is much smaller than that in the primary loop.

The primary reason for using chilled water for cooling equipment is cost. The systems are reliable, low in cost, small in size, readily available in many different sizes from many different

manufacturers, and easy to maintain. Air-to-air heat exchangers can, in some cases, be substituted for chilled water cooling, but are larger in size and will not work in some applications. Refrigerated cooling systems are more expensive, but operationally eliminate the need for disposing of tritium-contaminated water generated by the chilled water systems. The coolant may also become contaminated with tritium under some circumstances.

#### **4.5 Seismic and Other Natural Phenomena Design and Evaluation of Structures and Facilities**

A discussion of the DOE NPH requirements is included in Section 5.7.1. These requirements apply to the facility Safety Structures, Systems and Components (SSCs). Section 5.7.2 and its references should be reviewed as part of the overall NPH design of a new, or evaluation of an existing, tritium facility.

An understanding of the types of loading produced by earthquakes, extreme winds, and other NPH is useful in planning mitigating approaches. An earthquake produces vibratory ground motion, which will affect the entire facility and its contents. Attention to anchorage and connection details to all structures, systems, and components is essential. Earthquakes may also cause ground displacement if the facility is near a fault. Loss of ground stability may also occur due to settlement or liquefaction and will depend on soil types and location of the ground water table.

Extreme straight-line winds or tornadoes generally affect the structural shell and systems and components outside of the structure. Extreme winds will produce direct pressure and suction on walls and roofs with increased loading at corners, eaves, and ridges. Extreme winds can produce missiles from debris near the facility or from nearby facilities. These missiles can strike walls and roofs and their effects should be evaluated. During tornadoes, in addition to pressure effects and windborne missiles, there will be an atmospheric pressure change or a pressure drop below ambient pressure as the tornado passes over a facility. This can affect wall and roof openings and ventilation system filters. Flooding or extreme precipitation events can also have a deleterious effect on structures and safety SSCs, as discussed in DOE-STD-1020-2012.

An approach used at SRS is to store resources in a Highly Invulnerable Encased Safe (HIVES). HIVES's are metal cabinets designed to store tritium reservoirs and Hydride Storage Vessels (HSVs). The HIVES is designed to be capable of protecting the pressure boundary integrity of stored reservoirs or HSVs against impact of structural elements freefalling from the collapse of the H-Area Old Manufacturing Facility and the adjacent stack caused by design basis NPH events. A HIVES is constructed of hardened T-1 steel, primarily from 1-inch plate. The overall dimensions are approximately 22" W x 39" D x 68" H, which includes both a cabinet and a matching bonnet assembly bolted to the cabinet top plate. This bonnet contains an aluminum hexagonal cell honeycomb material designed to absorb the impact loading stated above.

The process for determining the NPH Design Category (NDC) for facilities and individual SSCs is described in Section 2.2 of DOE-STD-1020-2012. Not all portions of a facility, or SSCs in a facility,

must have the same NDC. NDC is determined by the radiological consequences of individual SSC failure. Once NDC is determined, Chapters 3-8 of DOE-STD-1020-2012 provide direction for determining the appropriate design basis loads for the various NPH. These chapters also provide design criteria, or reference national consensus standards for the criteria, to withstand the design basis loads.

Employing mitigation efforts before natural hazards occur is a very cost-effective means to provide life safety, to minimize damage and losses, and to reduce the impact on the facility and operations. It is extremely important to pay attention to all details because natural phenomena will find the weak links and cause damage.

#### 4.6 Other Design Considerations

The following items should be considered in the design of a facility with tritium operations.

- Designated safety-class systems should employ the concepts of redundancy, separation, and diversity. Some designs in the past, notably at the SRS H-Area New Manufacturing Facility, employed redundant signals to a single actuation device. An improvement to this design concept would be to provide these redundant signals to two independent, separate (by distance) actuation devices.
- The maintainability and accessibility of components is sometimes overlooked in the original design. Inaccessibility of glovebox components can result in serious ergonomic injuries to workers. Most current glovebox designs are not designed for ease of large equipment replacement, resulting in long and usually open-box maintenance. SRS is developing a modularization of tritium component concept that allows for reduced maintenance costs and schedule. Additionally, this modularization concept results in lower glovebox volume-to-surface-area, reducing glass, gaskets, and gloves; thereby producing the added benefit of reduced hydrogen loading into the glovebox, as the majority of hydrogen loading is from the permeation of humidity through gloves and gaskets<sup>31</sup>.
- A design requirement should be included to prevent the formation of explosive mixtures during the handling, processing, and storage of tritium gas and other hydrogen isotopes.
- Calibrated tanks and associated piping that are used for pressure-volume-temperature measurements should have surface treatment of their interiors (e.g., electropolished and passivated) to allow accurate volume measurements.

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<sup>31</sup> Randy Davis, *A Plan for Modularization of Tritium Components*. presented at the 34<sup>th</sup> Tritium Focus Group Meeting, Aiken, SC, April 22, 2014

- Tritium process and handling systems should use, wherever possible, nonflammable hydraulic, lubricating, and cooling fluids.
- The designer should consider not using hydrogenous fluids where they might become contaminated with tritium.
- The designer should consider providing for the retention of firewater with subsequent monitoring prior to disposal for all buildings where there is tritium.
- Barriers should be provided to prevent damage to equipment and injury to personnel while performing testing operations that could produce missiles or blast pressures. These barriers should be designed using conservative and proven design principles, such as those of DOE/TIC 11268, *A Manual for the Prediction of Blast and Fragment Loading of Structures*.
- An independent air system should be provided for breathing air. It should have dedicated, oil-free compressors or pressurized cylinders of breathing air, and provide breathing air in all areas of the tritium facility where it may be needed for maintenance operations and/or personnel safety. Contamination of the air supply (e.g., from refrigerant leaks or air intakes) should be detectable at levels low enough so as not to pose health concerns.
- The design considerations of the Fire Protection System should include the following:
  - determining safety classification of structures, systems, and components (SSCs) of both fire detection and fire suppression systems;
  - consideration of unique fire sources (e.g., uranium beds used for tritium storage);
  - compatibility of fire extinguishing agents with the fire sources in a tritium facility; and
  - containing and handling requirements for expended firefighting agents (including water inventory) that may become contaminated with tritium.

#### 4.7 Lessons Learned

SRS Building 232-H, the H-Area New and Old Manufacturing Facilities, and the Tritium Extraction Facility; the WETF, located at Los Alamos National Laboratory (LANL); and the Tritium Research Laboratory (TRL) at SNLL are examples of facilities that were initially designed to perform tritium handling. Other facilities in use now were originally designed to do other work and have been retrofitted to perform tritium handling.

One of the major problems with the facility retrofit process is that the existing utilities such as ventilation, floor drains, gas supplies, and chilled water systems are shared with the adjacent non-tritium areas. The same ductwork, which is used to sweep released tritium from tritium operating areas, is used to provide ventilation for offices and non-tritium areas, and, as a result, tritium back

diffuses into the non-tritium areas through the shared ductwork. The same chilled water system used to cool the tritium-related equipment is used to cool the non-tritiated office spaces, and leaks of tritium-contaminated chilled water result in contamination of clean areas. The floor drains from the non-contaminated areas drain into the same system as the floor drains from the tritiated areas, and, through the drains, gases flow from one area to another.

Tritium facility utilities such as chilled water, compressed air, gas supplies, ventilation, floor drains, sink drains, storm drains, and stacks should not be shared with other non-tritium areas. Sharing these systems spreads tritium contamination to other areas, complicates day-to-day management of the facility, and impacts the transition process if the facility is transitioned to other use. The use of hazardous materials should be reduced or eliminated in the initial design stages of the facility, as it will likely lead to the generation of mixed wastes and increased decontamination and decommissioning (D&D) costs.

#### 4.7.1 SNLL Tritium Research Laboratory

Tritium operations have been terminated at the TRL, and the facility has been transitioned to other use. The problems encountered during transition of the TRL can serve as an example for facility designers of the future. The TRL was designed beginning in 1972 specifically to handle tritium, and a few changes to the initial design would have resulted in significant cost and timesaving during the transition process.

- The tritium removal system in the central glovebox had a supply and return manifold fabricated of six-inch diameter stainless steel pipe. The associated vacuum pump effluent manifold consisted of an all welded two-inch diameter pipe. These manifolds were approximately 200 feet long and extended down the central corridor of the building. The manifolds extended into each room from the corridor and were of all welded construction. During the transition process, special tooling had to be purchased to cut the six-inch and two-inch diameter pipe into sections small enough to be disposed of as solid waste.

Designing the system to include flanges and isolation valves at specific locations would have resulted in some cost savings during facility dismantlement and transition. However, since too many valves and flanges can increase the potential for leaks during operation, the installation points should be placed only at strategic locations.

- The floor covering installed in the TRL consisted of 12-inch tiles glued to the concrete floor. Both the floor tile and the adhesive contained asbestos that had to be removed during transition of the facility. Additionally, tritium-contaminated liquids were spilled on the floor during operation and leaked through the tile into the concrete below. The use of adhesives made of non-hazardous materials would have resulted in significant cost savings.

PPPL has suggested that sealing the concrete with a thick, hard finish epoxy paint prior to installation of any floor covering would mitigate the impact of tritiated liquid spills, although even epoxy will experience tritium permeation.

As untreated concrete can hold a large amount of tritium—and it is difficult to decontaminate once it is absorbed—the selection and application of an appropriate coating or paint is critical to eventual decontamination efforts. Studies are underway in Japan that detail the effectiveness of various coatings<sup>32</sup> SRNL has deployed a very effective decontamination process for contaminated concrete (and soil) similar to a barbeque pit-type arrangement<sup>33</sup>.

- The TRL was equipped with a recirculating chilled water system. The water-to-gas heat exchangers used the chilled water to cool the glovebox and vacuum effluent tritium removal system gases, vacuum furnace heat exchangers, glovebox temperature control systems, and in a variety of other tritium-related tasks. After a few months of operation, tritium contamination was found in the chilled water. In order to control the buildup of tritium in this system, the contaminated water was periodically drained from the system and replaced with uncontaminated water.

If the initial design had used water-to-water heat exchangers as barriers, the volume of contaminated water would have been minimized. If double-walled, gas-flushed, water-to-gas heat exchangers had been used; the contamination would have been significantly lower.

- The TRL used approximately 100 oil-type vacuum pumps. Oil-free vacuum pumps were not as common when the TRL was designed. The vacuum pump oil becomes contaminated with tritium during use. At some DOE locations, tritium-contaminated oil is regulated as a mixed waste. Handling the tritium-contaminated oil is a significant safety hazard to operating personnel and should be eliminated where practical.

If oil-free vacuum pumps had been used, the generation of mixed waste in the form of tritium-contaminated vacuum pump oil would have been eliminated as would the hazard associated with changing vacuum pump oil.

- The TRL was equipped with a ten-air-change-per-hour, single-pass, pressure-zone-controlled ventilation system. Some of the ductwork became contaminated and was removed and disposed of as solid low-level (radioactive) waste during the transition process.

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<sup>32</sup> Yuki Edao, *Measurement of Tritium Penetration through Concrete Material with Various Paints Coating*. Presented at the Tritium 2013 Conference, Nice, France

<sup>33</sup> Dennis Jackson et. al., *Thermal Removal of Tritium from Concrete and Soil to Reduce Groundwater Impacts*. Presented at the 34<sup>th</sup> Tritium Focus Group Meeting, April 22, 2014

Experience from SNLL has suggested that if the ventilation ductwork for room air ventilation had been separated from the ductwork used for high velocity air hood and glovebox ventilation, the contamination would have been easier to control during the transition process.

- The presence of hazardous materials in the form of asbestos and oil complicated the transition process. The materials used in fabrication and during facility operation should be reviewed, and, if possible, all hazardous materials should be eliminated.
- The initial design of the TRL included collection of wastewater from floor drains and sinks in two underground holding tanks so that the water could be checked for contamination before it was sent for disposal. The holding tanks were buried and could not be inspected. The wastewater drain system consisted of several hundred feet of buried drainpipe, which drained into the holding tanks. After a few years of operation, the underground tanks were replaced with holding tanks enclosed in a below-ground-level open concrete pit. If the tanks leaked, the concrete pit would contain the leak. This design also provided for inspection of the tanks for leaks. The buried, underground drain lines remained in place throughout the life of the facility.

Wastewater holding tanks and collection systems should be designed so that potential leaks can be contained and the holding tanks and drain system can be inspected periodically.

#### 4.7.2 SRS Old Tritium Extraction Facility

Between 1995 and 1997, a team of specialists performed D&D work on the Old Tritium Extraction Facility (232-F) at the SRS<sup>34</sup>. The work was conducted in six phases. Each phase presented challenges regarding the behavior of tritium, particularly tritium in equipment and structures of metal and concrete. A description of each phase is as follows:

- Phase I, Pre-Characterization/Isolation, was designed to identify the specific buildings, ancillary equipment, structures and premises to be cleaned, and isolate them in a way that avoided inadvertent contamination elsewhere in the complex;
- Phase II, Detailed Facility Characterization, included determination of the type and amount of actual contaminants and determined the location and type of disposal operations required. This phase was the most complex in terms of characterizing tritium contamination;
- Phase III, Decontamination and Dismantlement, involved removing hazardous materials such as PCBs, mercury, and lead, as well as radioactive constituents such as tritium and fission products. It also involved dismantling equipment and some structures;

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<sup>34</sup> DOE/SR-5000-510, "Tritium Facility Decommissioning, Pioneering Success at the Savannah River Plant," October 1997

- Phase IV, Demolition with Explosives, involved the removal and destruction of the facility and stack;
- Phase V, Waste Management, was a continuing process throughout all six phases. It involved the determination of how much of each contaminant was present at the site and where the various site contaminants would be disposed;
- Phase VI, Green Grass Restoration, restored the site to a usable, visually aesthetic entity.

Detailed characterization was one of the most challenging phases in dealing with tritium, and required an expert understanding of the behavior of tritium in porous and nonporous materials. For example, because tritium migrates to the subsurface of porous materials, volumetric characterization required core boring. This was not well understood initially and early characterization activities, which involved smear and scabbling techniques or wiping and scraping the surface of concrete, gave false (low) tritium contamination levels. In some instances, areas that were smeared clean began to reveal elevated readings following rain in-leakage. In addition, destructive testing for nonporous material, such as metals, was required to fully characterize volumetric tritium contamination.

DOE-STD-1120-2005, *Integration of Environment, Safety, and Health into Facility Disposition Activities*, provides guidance for integrating and enhancing worker, public, and environmental protection during facility disposition activities.

#### 4.7.3 Component Plugging at SRS Tritium Processing Facility

One of the tritium processing facilities at SRS experienced plugging of multiple components. The first indication was that the let-down filter on the compressor plugged; upon further inspection, fine particulate was found widespread in the facility, requiring mitigation and downtime. Sources of particulate contaminants included polymers used in compressor poppets, valve packing, valve seats, other components, and material introduced during maintenance such as lubricants (Krytox<sup>®</sup> and SWAK<sup>®</sup>), cleaning agents (Freon<sup>®</sup> and Vertrel<sup>®</sup>), along with dust and dirt from line break activities.

The general lessons learned captured by the SRNL investigators are germane to most tritium facilities and worth bearing in mind:

- wear and tear of components happens;
- tritium degradation of materials happens, and critical systems should be protected to the maximum extent possible;
- appropriate protective measures should be implemented (e.g., filters);
- particulate generation probably cannot be eliminated, but can be minimized; and

- particulate flow, migration, and accumulation should be considered in facility design<sup>35</sup>

#### 4.7.4 Tritium Compatibility Lessons

##### 4.7.4.1 Unanticipated Material Interactions

The damage done to organic materials by the presence of tritium in the internal structure of the material is not limited to the more obvious radiation damage effects. Tritium, particularly in the form of T<sup>+</sup>, has the insidious ability to leach impurities (and nonimpurities) out of the body of the parent material. In many cases, particularly where halogens are involved, the damage done by secondary effects such as leaching can be more destructive than the immediate effects caused by the radiation damage. In one such case, the tritium contamination normally present in heavy water up to several curies per liter was able to leach substantial amounts of chlorides out of the bodies of neoprene O-rings that were used for the seals. The chlorides leached out of the O-rings were subsequently deposited into the stainless steel sealing surfaces above and below the trapped O-rings, which led directly to the introduction of chloride-induced stress-crack corrosion in the stainless steel. The operational conditions that set up the introduction of the stress-crack corrosion were moderately elevated temperatures (i.e., less than 100 °C), low pressures (i.e., less than 3 atmospheres), and exposure times of 3-5 years. Fortunately, the damage was discovered before any failures occurred, the neoprene O-rings were removed, and the seal design was changed to a non-O-ring type of seal.<sup>36</sup>

In a second case, six failures out of six tests occurred when high-quality Type 316 stainless steel was exposed to tritium gas in the presence of Teflon<sup>TM</sup> shavings and 500 ppm moisture. All of the failures were catastrophic, and all were the result of massively induced stress-crack corrosion. The conditions that set up the introduction of the massively induced stress-crack corrosion in this case were moderately elevated temperatures (i.e., 104 °C), high pressures (i.e., 10,000 to 20,000 psi), and exposure times that ranged from 11 to 36 hours. Since the time to failure for all the tests was directly proportional to the pressure (i.e., the higher pressure tests failed more quickly than the lower pressure tests), since identical control tests with deuterium produced no failures, and since comparable testing without the Teflon<sup>TM</sup> shavings indicated no failures after 3,200 hours, it was concluded that fluorides were being leached out of the Teflon<sup>TM</sup> and deposited directly into the bodies of the stainless steel test vessels.<sup>37</sup> An interesting sideline to this test is that, after the tests, the Teflon<sup>TM</sup> shavings showed no obvious signs of radiation damage (i.e., no apparent discoloration or other change from the original condition).

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<sup>35</sup> P. Cloessner, SRNL, *Particulate Generation in a Tritium Facility*, presented at the 33<sup>rd</sup> Tritium Focus Group Meeting, Aiken SC, April 22-24, 2014.

<sup>36</sup> DOE HDBK 1132-99, Reaffirmed 2014, Design Considerations

<sup>37</sup> DOE HDBK 1132-99, Reaffirmed 2014, Design Considerations

#### 4.7.4.2 Valves with incompatible valve material procured, installed, and tested ready for use at a new SRS Facility

The author of this Standard was a member of a DOE/NNSA Operational Readiness Review (ORR) team pre-visit for a new SRS tritium facility in 2006, not because of his tritium expertise, but rather due to his responsibilities as the POC for the Office of Primary Interest (OPI) for DOE O 425.1 in the area of nuclear facility startup and restart; there were in fact no other tritium personnel on the ORR Team. The author, in 1993 was the lead for a review team for initial startup of a SRS tritium facility and not being convinced by WSRC's assessment of the capabilities of Dupont's ultra-high molecular weight polyethylene UHMWPE to withstand tritium environments without degradation, had negotiated with WSRC a periodic surveillance program and accelerated bench testing of the UHMWPE valves as one of the conditions for his team's recommendation for initial startup of this facility. Partly based on this experience, the author saw a need for DOE-wide guidance in the area of overall criteria for selection of valves in tritium service. He researched and published a number of articles<sup>38 39</sup> and a DOE guidance document<sup>40</sup> for selection of proper types and materials of valves for use in differing applications and varying degrees of tritium service. These publications include discussions of significant historical DOE tritium release events.<sup>41 42 43 44</sup> The number-one recommended practice in references 40 through 42 was "do not use halogenated elastomers or polymers such as Kel-F™ Teflon®, or Viton® for wetted parts in valves used for extensive tritium service." The author, being fluent in the language of the part numbers stamped on the bodies of the tritium valves was able, during the pre-ORR visit walking tour of the SRS facility in 2006, to identify inappropriate (e.g., Kel-F™) valves in the tritium systems. The ½" BK series Nupro® valves contained PCTFE (polychlorotrifluoroethylene) stem tips and a PTFE (Teflon®) coated bellows gasket, which are incompatible with tritium service, and were being used throughout the facility violating the number-one recommendation above.

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<sup>38</sup> W. Weaver, *Guidelines for Valves in Tritium Service*. Fusion Technology July 1994.

<sup>39</sup> W. Weaver. Selection Criteria for Valves used in Tritium Service. Presented at the International Workshop on Physics and Technology of Tritium for Fusion Reactors, Varenna, Italy, 6-14 September 1993.

<sup>40</sup> W. Weaver. EH Technical Notice 94-01, *Guidelines for Valves in Tritium Service*, 1994.

<sup>41</sup> *Tritium Release During Downsizing Activity (B-331)*, Occurrence Report SAN-LLNL-LLNL-1991-1002, U.S. Department of Energy, April 1991.

<sup>42</sup> *Tritium Release – Equipment Failure*, Occurrence Report ALO-KO-SNL-TRL-1992-0002, U.S. Department of Energy, June 1992.

<sup>43</sup> *Environment Safety & Health Safety Note*, 91-3, U.S. Department of Energy, March 1992.

<sup>44</sup> *Tritium Release Not Part of Normal Operation*, Occurrence Report ALO-LA-LANL-TSTA-1991-0065, U.S. Department of Energy, March 1991.

The subsequent investigative review<sup>45</sup> performed by the Contractor also identified that these same valves were procured and were being used as spares for the operating facilities. Fortunately none of the 105 spares had yet been placed in service in the operating facilities and tritium had not yet been introduced into the new facility with its 195 installed valves. A thank-you call<sup>46</sup> from the Federal Project Director of the new facility to the author indicated that he had averted a major disaster by identifying the issue before tritium was introduced.

This event highlights the need for technically competent independent reviews, which is a recurring theme across the DOE complex. Both aspects, technical competence and independence are required for success. Too often, management (DOE and Contractor) views safety or independent technical reviews as an impediment to progress and press on without adequate independent technical reviews. It is rare that these reviews are detrimental, as they are neither costly nor time consuming, and are usually able to identify issues if they exist, before they become problems. One need only to look at the lack of early identification of the multiple design issues at the Waste Treatment and Immobilization Plant (WTP), the technical design problems delaying startup at the Integrated Waste Treatment Unit (IWTP), the undiscovered design errors at Uranium Processing Facility (UPF) and the LANL/WIPP waste packaging issue to illustrate the need for better independent technical reviews. For example, during the packaging of waste containers at LANL for disposal at WIPP, the procedure called for adding chemicals that were not compatible within the same container, which might result in a potentially reactive mixture. During the interview process at LANL, organic chemists stated to this author, who was part of the accident investigation of the WIPP radiological release event of February 14, 2014<sup>47</sup>, that if they had been involved in the review process of the packaging process, they would have identified this incompatibility issue in the procedure as a poor technical practice, independent of any potential consequences. The cost associated with this type of review would have been minimal. Additionally, the Inspector General (IG) Management Alert of September 2014<sup>48</sup> further identified a lack of adequate technical review as a weakness in the LANL process. Likewise, the Tritium Focus Group discussed that the author's pre-ORR walkdown was probably the least amount of time expended for the most amount of money saved in the history of the DOE complex. While this may indeed be true, the technically competent reviewer has invested much time to achieve the expertise and should be used to the fullest extent possible. However, a recurring theme seen in these major mishaps is management's lack of awareness and use of technical personnel available. Even in the example of the success of the author's pre-ORR identification, it was due to his OPI function for the ORR and not tritium expertise that he was involved with the ORR team. Shortly after returning to Headquarters from the pre-ORR visit, his organization was dissolved, and as such he was no longer involved with OPI activities for the

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<sup>45</sup> ORPS Report NA--SRSO-WSRC-TRIT-2006-0002, *Incompatible Nupro Valves Stem Tip Material*, finalized 09/28/2006.

<sup>46</sup> Personal communication from Kevin Hall to Bill Weaver, August 15, 2006.

<sup>47</sup> DOE EM Accident Investigation Phase II Report, Radiological Release at the Waste Isolation Pilot Plant, April 2015.

<sup>48</sup> Management Alert, US DOE Office of the Inspector General, DOE/IG-0922, September 2014.

ORR. Although he was the only Federal employee in the NNSA/DOE complex with tritium in his job title, he was slated for transfer to an organization that had little to no involvement in tritium activities, and was removed from the ORR team.

#### 4.7.5 Oxygen monitor failures

Older oxygen monitors have been observed to be failing at a SRS Tritium facility and spare board are difficult to obtain. These boards are the essential components for the Delta F analyzers. The Tritium facilities currently have 24 of these analyzers that are used to monitor the Oxygen levels in the glovebox atmosphere. These analyzers are credited Safety Significant controls and their purpose is to ensure the glovebox oxygen concentration is below the minimum concentration required to support combustion inside the glovebox.

Since the Delta F analyzers are over twenty years old, the manufacturer no longer supports this analyzer and spare parts are no longer available. Until the upgrading of the facility's oxygen monitor systems is complete, the Delta F system must remain operational. Hence, the option to repair the circuit boards was pursued.

During the repairs, it was discovered that the opamps in the cell amplifier and x100 amplifier were the most common failures. A pattern developed indicating that LTC1500 and MAX430 opamps were failing. Due to the location of these opamps, it is suspected that the age of the chips is at fault. Since these chips are not in production, surplus parts were used to replace failed chips. The replacement opamps also fail on a regular basis.

To bolster the spare parts program, a project was funded to reverse engineer and redesign the Delta F main board. The redesigned board will significantly reduce the failure rate and allow the normal process for instrument replacement to continue.

#### 4.7.6 Tritium flammability limits in glovebox atmospheres.

Los Alamos National Laboratory declared a Potential Inadequacy of the Safety Analysis (PISA) documented in the September 17, 2014, Occurrence Reporting and Processing System report, NA-LASO-LANL-TRITFACILS-2014-0006<sup>49</sup>. This PISA was due in part by the fact that hydrogen species (including tritium) have a lower flammability limit in argon/air mixtures compared to air alone.

The presence of inerts (argon, nitrogen) changes the hydrogen lower flammability limit (LFL) and changes the limiting oxidant concentration (LOC). The Combustion Research Center (CRC) 2006 publication "Flammability Limits of Hydrogen – Air – Argon and Hydrogen – Air – Nitrogen Mixtures" reported a hydrogen LFL of 3.68% (~3.7 vol. % hydrogen, 26.8 vol. % air, and 69.5 vol. % argon) and

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<sup>49</sup> Occurrence Reporting and Processing System report, NA--LASO-LANL-TRITFACILS-2014-0006, September 17, 2014

an oxygen LOC of 3.33% (~6.5 vol. % hydrogen, 16 vol. % air, and 77.5 vol. % argon)”<sup>50</sup>. Per the CRC 2006 publication, nitrogen environments yielded a higher LFL (4.44% hydrogen) and higher LOC (4.87%) compared to argon environments. As discussed in section 4.1.1.a(2)(b), *Medium-Quality Secondary Container*, tritium gloveboxes in the DOE complex are operated at a slight negative pressure to the room and could experience air in-leakage resulting in the Hydrogen-Air-Argon or Hydrogen-Air-Nitrogen atmospheres such as those examined in the CRC.

Other glovebox atmospheres such as argon-hydrogen-oxygen, which are atypical for DOE, have been shown to have LFLs that are lower than for argon-hydrogen-air environments<sup>51</sup>. An experimental data point<sup>52</sup> indicates a lean flammability limit below the 3.68% CRC 2006 value although details are lacking at this time to correlate these findings with LFL and the lean flammability results are not in agreement with published LFL data. For example, a report from Fauske & Associates<sup>53</sup> states: “for the case of air ingress into an existing H<sub>2</sub>-Ar mixture, a lower oxygen limit between 4% and 5% is recommended by using a linear fit:  $x_{O_2} = 5 - (x_{Ar}-50)/20$  where x is mole percent and  $50\% < x_{Ar} < 70\%$ . Use 5% O<sub>2</sub> when  $x_{Ar} < 50\%$  and use 4% O<sub>2</sub> when  $x_{Ar} > 70\%$ . However, the mixture is completely inert when  $x_{Ar} > 76\%$ .”

Other publications<sup>54</sup> test mixture flammability inside a long tube instead of inside a pressure vessel as specified by ASTM standards. Flame propagation inside a long vertical tube ignited from the bottom is aided by natural convection. These experiments therefore produce more conservative flammability results (i.e., broader lean and rich flammability limits). The applicability of these results vs. pressure vessel flammability results for gloveboxes needs to be determined.

Not all reported results in this area are of the same quality or applicability. The CRC data has the highest pedigree and is the best to reference. It has the advantage of using a standardized accepted test method (ASTM) and better instrumentation compared to early tests on flammability. Tests were reproduced and thus the accuracy of the data is available. The Fauske report uses data from the Van Heiningen report which was issued in 1936. The apparatus used for the 1936 study involves vertical tubes and potential for hydrogen buoyancy as discussed above. Additionally, the Van Heiningen report uses several different references for work done even earlier. Data is reported to one decimal point; no indication is provided on reproducibility. The CRC study reported values measured to two decimal points and generated many more data points to determine the flammability curve. The early studies used oxygen instead of air whereas the CRC studies used air.

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<sup>50</sup> Combustion Research Center, Flammability Limits of Hydrogen – Air – Argon and Hydrogen – Air – Nitrogen Mixtures, Report No. CRC-2709 Rev. 3, Issued May, 9 2007

<sup>51</sup> Shebeko, Y. N., *et al.* “The Influence of Inert Retardants on the Combustion of Hydrogen-Oxygen Mixtures under Elevated Temperatures and Pressures”. *Combustion, Explosion, and Shock Waves*. Plenum Publishing Corp., Vol. 30, No. 2, 1994

<sup>52</sup> Terpstra, M. A. “Flammability Limits of Hydrogen-Diluent Mixtures in Air”, University of Calgary, 2012

<sup>53</sup> Flammability and Combustion of H<sub>2</sub>, Air or O<sub>2</sub> and Ar Gas Mixtures, M.G. Plys, Attachment 11 to SNF-22059, Rev 0 July 2004

<sup>54</sup> Coward, HF and Jones GW, Limits of Flammability of Gases and Vapors, Bulletin 503, US Bureau of Mines, 1952

Because of the questions raised in citing old data from questionable test apparatus, instrumentation that is not as precise as what is available today, and use of oxygen instead of air, these results have less credibility than the CRC values.

Safety analyst and operations personnel associated with tritium gloveboxes using argon or nitrogen should be aware of the potential adverse impact on LFLs and LOCs based on their specific conditions and configurations.

#### 4.7.7 Pantex Tritium Release from Gas Reservoir

Pantex experienced a large tritium release in 1989. This and other releases across the DOE complex concerned Secretary Watkins to the point that he commissioned an eight member Tritium Task Group (TTG) in 1991, to visit every DOE tritium facility and review not only the releases but also to provide him with insight on how to improve operations. This author was one of the eight TTG members, three of which are still active DOE employees. There was also a thirteen member Working Group available for support to the TTG. The current Tritium Focus Group (TFG), which now meets semi-annually and whose Charter is Appendix H, was initially formed to respond to the findings and recommendations of the TTG. Only one unintentional tritium release has occurred at the Pantex Plant in its 40-year operating history. The incident occurred on May 17, 1989, during a normal weapons disassembly and retirement operation. An electroexplosive-actuated valve operated accidentally, resulted in discharge of tritium from the gas reservoir into the disassembly cell, and ultimately to the environment. The incident resulted when the disassembly technician, who did not realize the valve had malfunctioned, loosened a gland nut on the reservoir. The equipment contaminated by this event continued to offgas tritium, which accounted for the majority of the tritium release from the site in fiscal year 1990.

When the tritium was released into the cell, portable and fixed tritium monitors alarmed and all workers immediately exited from the cell. Of the five workers in the area at the time of the incident, four received negligible exposures while the fifth worker received a significant dose.

As a result of this incident, three investigation teams were assigned to examine various aspects of the event. The first team evaluated the cause of the accident, assessed the nature and extent of the tritium release, personnel exposures, and Plant contamination, and determined the adequacy of the Plant's emergency response at the time of the incident. A second team evaluated the design characteristics of all electroexplosive devices in the weapons stockpile and assessed weapons handling procedures at the Pantex Plant. The third group reviewed tritium operations at the Pantex Plant and the associated airborne effluent monitoring, filtering, and confinement systems.

The purpose of the third team's review was to identify options for improving the safety of tritium operations and to determine associated costs and implementation schedules. The review focused on what actions could be taken at the Pantex Plant to eliminate or mitigate the consequences and effects of tritium release on assembly and disassembly workers, other Pantex Plant employees, the general public, and the environment.

As a result of the first team's investigation and recommendations, changes were made in the Pantex Plant disassembly operations procedures. The operators now monitor for tritium prior to and during the tritium reservoir removal. Additionally, improvements were made in operator radiological training and in Plant operating and emergency procedures, and the health physics staff was increased.

The second investigation team recommended modifications to the design characteristics of electroexplosive devices in the weapons stockpile and revisions to the Pantex Plant weapons handling procedures.

The third investigation team identified four improvement options in equipment and facilities, and five improvement options in environmental monitoring and protection.

## 5.0 DESIGN OF EQUIPMENT

The design requirements for tritium are a function of the tritium form, quantity, concentration, pressure, and period of storage. High concentrations of tritium gas stored at high pressure ( $> 2,000$  psia) are difficult to contain due to tritium and helium embrittlement of the container materials. Design of these systems requires careful selection of the materials of construction and should be designed using expertise in high-pressure tritium containment.

Low concentrations of tritium in gaseous form mixed with other gases at low ( $< 600$  psia) to medium (600 to 2,000 psia) pressure, regardless of the quantity, do not significantly impact the strength of the materials they are stored in. As a result, standard designs can be used.

Tritiated water in the form of  $T_2O$  is somewhat corrosive unless properly stored with an overpressure of  $T_2$  gas. This is due to the suppression of the formation of oxygen in the cover gas and peroxides in solution<sup>55</sup>. Tritium systems should be designed by persons with tritium experience.

Low concentrations of HTO (mCi/mL to Ci/mL) recovered from tritium removal systems have proven to be corrosive when stored in liquid form in metallic containers and have resulted in the development of significant leaks in containers within days or weeks. Storage of this same water solidified on clay or on molecular sieve material, regardless of the quantity, is stable and noncorrosive and may be stored for many years in the container.

### 5.1 Material Compatibility

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<sup>55</sup> John Gill, Babcock & Wilcox of Ohio, Personal Communication to W. Weaver, July 1998.

Proper materials selection and rigorous design have led to tritium-handling systems that are extremely safe for long periods of time. Materials exposed to tritium under certain conditions can be susceptible to hydrogen embrittlement. The chances of embrittlement are significantly reduced by proper material selection.

Tritium can permeate vessel barriers, especially in components operating at elevated temperature. Currently available tritium permeation data are normally sufficient to estimate the order of magnitude of tritium permeation through barriers. These estimates can be used during system design or to determine whether additional purging and stripping systems are required to “clean up” permeated tritium or whether other design changes (such as wall thickness, material, or coating) are required to reduce permeation. Tritium permeation can lead not only to contamination outside the barrier, but it can also result in significant quantities of tritium being dissolved in parts, which can lead to hydrogen embrittlement. Over time, this tritium decays to  $^3\text{He}$ , which has been found to accentuate hydrogen embrittlement and to cause weld cracking (see below).

#### 5.1.1 System Design

High quality tritium primary containers should have a low leak rate and probability of failure or leaking. The consequences of tritium leaks can include personnel uptake, release to the environment, ignition (if mixed with oxygen), and violation of operating permits. Pressure and vacuum vessels used in tritium systems are generally designed and constructed using codes and standards applicable to boiler and pressure vessels. In the United States, use of the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code is recommended but not required. Pressure and vacuum vessels constructed using this code have an accepted rigor in design, construction, and inspection that facilitates approval and acceptance by regulators. The ASME Boiler and Pressure Vessel Code primarily cover the design of vessels, but does not cover all of the aspects of a tritium system design. No real tritium compatibility specifications exist for tritium relief devices, and ASME leak rate standards are only in the  $10^{-3}$  leak rate region. Other design standards, such as resistance to seismic events, also are to be followed, depending on the location and regulators of the facility. See Sections 4.5 and 5.7 for more discussion on seismic design.

#### 5.1.1.a Leak Testing

After fabrication, thoroughly leak-testing tritium systems are extremely important. Normally, leak testing employs commercial helium-mass-spectrometer-based systems and is performed after any other required proof, pressure, or vacuum performance tests. Other leak detection methods, such as rate-of-rise, can also be employed in addition to helium mass spectrometry. A dilute solution of tritium in an inert gas can also be used to detect small leaks. Tritium is a highly effective leak detection species, since it travels rapidly through cracks and can be easily detected at very low levels.

#### 5.1.1.b Joining

Components of tritium systems are commonly joined by welding. Welds are normally designed so they can be non-destructively inspected by a suitable method such as radiography or ultrasonic testing. Also, the weld design should minimize so-called “virtual leaks” on the interior of tritium containing volumes. Examples of weld practices to minimize virtual leaks include 1) using full penetration welds where possible, and 2) welding feed-through to the interior wall surface (not on the outside, which would leave a gap on the inside around the feed-through that is difficult to outgas). Standard weld rod filler materials are chosen, depending on the base alloy. Every effort should be made to reduce or eliminate residual welding stresses. Oxide-free welds are also preferred to decrease tritium holdup due to tritium-protium exchange on the oxide. The welds can be produced by inert gas flow through tubing and piping or by wire brushing using a suitable alloy wire brush.

There is less experience using other joining methods such as brazing or high temperature soldering in tritium systems. Dissimilar materials may have to be joined by a transition junction, using an intermediate material to enable proper welding and accommodate differences in thermal expansion and other properties.

Tritium gas permeates austenitic stainless steels, and, over time,  $^3\text{He}$  is created by beta decay of tritium in solution in the material. Welding stainless steel containing solute helium is difficult because intergranular cracking can occur. During welding, the solute helium agglomerates at grain boundaries and forms both intergranular cracks in the heat-affected zone and pores in the fusion zone. Low-heat-input weld techniques have been shown to mitigate this problem to some degree; however, weld repair of helium-containing stainless steel is normally difficult to perform without some cracking.

All-metal mechanical joints are also a sound way to join components in tritium systems. Typically, copper, silver-plated nickel, or silver-plated stainless steel have been used as gaskets. Commercial high and ultrahigh vacuum fittings are normally compatible with tritium.

### 5.1.1.c Surface Coatings and Treatments

Aluminum and aluminide coatings have been successfully employed on stainless steel to reduce permeation into and through the steel. These coatings can be applied on large items using a proprietary fluidized bed furnace, having a controlled atmosphere (in the so-called “calorization” process), using a suitable aluminum-rich slurry coating, employing chemical vapor deposition, or a “pack bed” process. Gold has also been used as a permeation barrier in some applications and is often applied over a thin nickel buffer layer (“strike”) that has been applied to the bulk metal (e.g., stainless steel) after proper surface preparation.

Several companies treat stainless steel surfaces using various proprietary electrochemical processes to “passivate” the surface. These processes probably enrich the chrome content of the surface oxide, polish the surface (thereby reducing the effective surface area), and remove carbon and hydrogen from near the surface. All of these microstructural changes may be desirable for tritium systems in which the process gas must remain at high purity. Capillary lines that route gas to mass spectrometers are commonly passivated to reduce changes of gas composition by isotope exchange, while the gas flows from the sample location to the mass spectrometer. Passivated surfaces reduce the rate of isotope exchange in hydrogen isotope mixtures, probably by reducing the catalytic effect of the surface decomposing hydrogen isotope molecules to atoms, which enables isotope exchange. Vacuum systems having surfaces treated in this way evacuate faster. This type of surface passivation can be expensive; so many parts of tritium systems are not passivated; normally only parts requiring the special properties of passivated surfaces are treated.

### 5.1.2 Structural Metals

Exposure of metals to high pressure ( $> 2,000$  psia) hydrogen, deuterium, or tritium may result in hydrogen embrittlement of the material. This could eventually result in material failure. The time until failure is a function of the container material, the pressure, and temperature. Additionally, materials exposed to high-pressure tritium are also subject to helium embrittlement. Tritium at high pressure enters the metal and decays to  $^3\text{He}$ . The buildup of helium in the metal results in helium embrittlement, which, depending upon the pressure, temperature, and type of material, will eventually result in failure of the material. Exposure of metals to low- to medium-pressure tritium at normal temperatures does not generally result in material failure within any reasonable period of time.

Some metals are more resistant to embrittlement than others and, therefore, are more compatible with tritium. Depending upon the specific application, Types 304L and 316L stainless steel are generally considered to be the most hydrogen compatible and readily available stainless steels for tritium service. High-pressure vessels, valves, and tubing designed of these materials when used at their rated pressure and temperature will provide many years of service without material failure. When equipment is designed for tritium operations, a materials expert should be consulted to ensure that the materials selected are compatible with their intended service.

### 5.1.2.a Austenitic Stainless Steels

The recommended materials of construction for tritium-handling systems are from the class of wrought 3XX series of austenitic (face-centered-cubic) stainless steels, including Types 304L, 316L, and 347. Types 304L and 316L stainless steel are most often used in tritium processing systems, but are limited to temperatures less than 425 C. These steels provide good strength, weldability, and resistance to hydrogen embrittlement. Components fabricated from these materials are procured routinely. Many commercially available vacuum system components that are used in tritium systems, such as valves, piping, pumps, and analytical instrument sensors, are fabricated from these types of austenitic stainless steel. Wrought materials are preferred to cast because wrought materials normally have a more homogenous microstructure. In the past, tritium has leaked through parts having poorly oriented stringers and inclusions, including seamless tubing. The forging direction of some wrought components has been specified so that the orientation of inclusions is not in a direction that could result in a tritium leak path. Low carbon grades (such as 304L and 316L) are preferred to avoid weld sensitization and to reduce the number of inclusions (impurity particles such as oxides and carbides). Modern vacuum-arc-remelted steels are a good choice because they have lower impurity levels, thereby resulting in fewer inclusions that could aid hydrogen-induced cracking or provide leak paths. Typically, tritium system components employ seamless pipe and tube where practical.

Stabilized grades, such as Type 347, have been employed in applications where post-weld heat treatment is not possible. This usually occurs when a process vessel contains a working material (such as hydride or getter) that will degrade when exposed to the post-weld heat treatment, which is typically performed at about 1,100°C for austenitic stainless steels.

High carbon grades, such as Type 347H or Type 316H (having 0.04 percent carbon minimum), have been successfully employed for tritium service if high temperature strength is required. Type 347H is employed in the Hydride Transport Vessel (see section 6.2.2), and Type 316H was employed for the SRS Building 232-H Extraction Furnace retorts. Type 310 stainless steel has good oxidation resistance and can be considered for elevated temperature applications if oxidation is a concern. Type 316 stainless has superior creep resistance but inferior oxidation resistance to Type 310.

Some types of higher-strength austenitic stainless steels not generally employed for tritium service may be required for fasteners (such as nuts and bolts) in mechanical joints in high-temperature regions. This may be acceptable if the bolts are exposed to only residual amounts of tritium. These materials also may be used to contact 3XX stainless steels to avoid galling of mating screw contact surfaces. Typical materials used in these applications include Nitronic 60, Nitronic 50 (also called 21-13-9) and Nitronic 40 (also called 21-6-9); these are all nitrogen-strengthened austenitic stainless steels.

### 5.1.2.b Copper and Copper Alloys

In principle, copper is a suitable material in tritium systems. Copper has several tritium-compatible properties. Tritium has a low permeability in copper, and copper is a ductile, stable, face-centered cubic metal and so is resistant to hydrogen embrittlement. The high thermal conductivity of copper is a desirable property for process vessels requiring heat flow or constant temperature. Copper can be easily joined in a number of ways (e.g., soldering, brazing, and welding). In spite of these advantageous properties, copper and copper alloys are not commonly used in tritium systems. Several factors may account for this. The ASME allowable design strength of copper falls rapidly at temperatures above 200°C, making it difficult to use copper at elevated temperature. Also, hydrogen isotopes can react at elevated temperature with oxygen in copper, whether the oxygen is in solid solution or in copper oxide precipitates. In either case, water is formed and over time water vapor agglomerates at grain boundaries, which eventually results in intergranular cavitation, cracking, and failure. This failure mechanism is sometimes termed “steam embrittlement.” Also, a transition junction (normally nickel) is required to join copper and the stainless steel components of the remainder of the system.

### 5.1.2.c Aluminum and Aluminum Alloys

Aluminum has properties making it potentially desirable for tritium systems. It has a low density and a high thermal conductivity. Hydrogen isotope permeability is very low in aluminum compared to stainless steel. Aluminum is used in applications where light weight is important, such as in containers that must be lifted by personnel in gloveboxes. Aluminum can be used for the construction of medium-quality static containment vessels. However, aluminum is not commonly used in tritium systems. Stainless steel has much higher strength, at room and elevated temperature. Welding aluminum requires more precautions because aluminum reacts with atmospheric water vapor, which can cause porosity due to hydrogen in the weld fusion zone.

### 5.1.2.d Materials to Avoid

Plain carbon steels and alloy steels cannot be used for tritium service. These steels have high strength and (normally) a body-centered-cubic crystal structure, both of which make the material less ductile and much more susceptible to hydrogen embrittlement. Ferritic stainless steels (such as Type 430), martensitic stainless steels (both quench-and-tempered (such as Type 410) and precipitation hardening (such as 17-4 PH and PH 13-8 MO)) and precipitation hardened austenitic stainless steels (such as AM-350) should not be used for general tritium service; they are all more susceptible to hydrogen embrittlement than the austenitic stainless steels. Additionally, free-machining grades of austenitic stainless steel (such as Type 303) should not be used.

Other materials that cannot be used for tritium service are any material that forms hydride near room temperature and atmospheric pressure. Examples include zirconium, tantalum, niobium, and many alloys of these materials.

Powders should not be used in or on containment and confinement systems. Materials in powder form and materials that are prone to form powders, such as insulation cloth, fabrics, polymers, and ceramics should be carefully considered and avoided if possible.

### 5.1.3 Polymers

All polymers degrade when exposed to radiation. Both tritium and tritiated water permeate all polymers, and permeated tritium deposits the beta decay energy throughout the polymer bulk. (Although the tritium beta energy is very low and has a small penetration depth in matter, permeation allows tritium atoms to be near enough to polymer chains throughout the bulk to cause changes in the polymer by radiation.) Types of radiation-induced changes in polymer properties include softening (degradation) or hardening, ductility loss, color change, dimensional change, and gas evolution. Because of these effects, polymers should only be used in tritium systems where no metal alternatives exist. Normally, only polymers that harden during radiation exposure are employed, and are replaced before they begin to deteriorate. In addition to polymer breakdown itself, products of degradation can form corrosive liquids or acids such as HF and HCl. Polymer parts have to be easily replaceable as a part of normal operations, and a program of regular inspection and replacement should be established. The system should be designed to expose any polymers to as little tritium as possible. Typical uses of polymers in gas-handling systems include gaskets, O-rings, electrical cable insulation and valve parts, including seats, stem tips, and packing.

Polymers relatively resistant to radiation can typically withstand up to about 1 million rad (1 rad = 100 erg energy deposited per gram of material). By knowing the solubility of tritium in a polymer at a given temperature and tritium partial pressure and the decay rate of tritium, the approximate dose can be calculated, assuming the tritium concentration has reached equilibrium.

Many effects of radiation on polymers are accentuated by oxygen. Protecting polymers from oxygen or air will likely lengthen the lifetime of polymers exposed to tritium. Also, temperatures above about 120°C accelerate radiation effects in polymers, so the temperature of any polymer parts should be kept as low as possible. Inert additives such as glass or graphite generally enhance the resistance of polymers to radiation. Addition of antioxidants may also enhance radiation resistance.

#### 5.1.3.a Plastics

Vespel®, a polyamide, has been successfully used for valve stem tips in some tritium laboratories. Ultra-High-Molecular-Weight Polyethylene (UHMWPE) and High-Density Polyethylene (HDPE) have been used for valve stem tips in automatic valves at SRS. Although the latter two materials were designed by DuPont™ specifically for tritium service at SRS, their performance has not reached the desired level. Use of HDPE has been discontinued. Valves that use UHMWPE are selectively transitioned to Vespel if the service conditions warrant the change and as the need arises to repair

or replace. EH Technical Notice 94-01, *Guidelines for Valves in Tritium Service*, describes in detail differing materials and actuators for varying applications. Low-Density Polyethylene (LDPE) is very permeable by tritium and tritiated water and should not be considered for use in tritium systems. Polytetrafluoroethylene (PTFE), trade-named Teflon®, degrades and decomposes in tritium, resulting in the formation of HF. In humid air, HCl and HF are formed, which are both highly corrosive. Generally, chlorofluorocarbon polymers should not be used in tritium service. Polyvinyl chloride (PVC) and vinylidene chloride (Saran™) are among several polymers used in tritium protective clothing, but should not be used for process equipment because they contain chlorine.

#### 5.1.3.b Elastomers

Tritium readily permeates into and diffuses through elastomeric materials and, depending upon thickness, begins appearing on the outside of the elastomeric seal within hours after exposure. Elastomers are subject to radiation damage. They harden and lose their sealing ability due to exposure to high concentrations of tritium.

Ethylene propylene diene monomer (EPDM) elastomers are employed for low-pressure process flange gaskets because of EPDM's relatively good performance in tritium service. In some cases, Buna-N process flange gaskets are being replaced by EPDM when the gaskets are changed; however, in other applications, Buna-N remains in use. Viton®, a common O-ring material, is used, but can embrittle in months in tritium service. Butyl rubber has low permeation for both tritium and tritiated water, but is not as resistant to radiation damage as EPDM. Butyl rubber is used for glovebox gloves. Silicon rubber can be used in glovebox construction and has been found to be effective at Mound and other sites. Water vapor in the air outside gloveboxes permeates gloves and can lead to a significant portion of the residual tritiated water vapor in tritium gloveboxes.

Kel-F®, and its successor Neoflon™, are common chlorofluorocarbon polymers (Polychlorotrifluoroethylene (PCTFE)) and are incompatible with tritium. They, like Teflon®, degrade in tritium gas and should not be used.

#### 5.1.4 Fabrication Cleaning and Testing

Materials used in tritium should be fabricated in a manner that produces little contamination. Methods that are consistent with high vacuum processes are often useful. When joining is required, gas tungsten arc welding (GTAW) is the preferred welding method. Other joining processes; e.g., gas metal arc welding (GMAW), tend to be prone to pore formation and thus are not suitable for storage vessels.

As indicated previously, forged materials that are designed to orient stringers, inclusions, and properties favorably are preferred to cast structures. Also, thin wall tubing may require additional testing, such as pressurized proof testing, to ensure suitability for service. SRS has experienced tubing failures due to wall stringers oriented parallel to the tubing. These leak sites can be weld-repaired.

Welds should be purged to prevent oxide formation or should be cleaned with a wire brush to remove any oxide film present. If stainless steel materials are wire brushed, the brush should be made from stainless steel and should not be used on carbon steel or stored in an area where inadvertent contact can be made with stainless steel products.

Any surfaces that require cleaning should be cleaned with compounds that contain little to no halide compounds. Halides are to be avoided since they can promote stress corrosion, cracking, and leaks. Solvents used in gloveboxes should clean effectively, but not contribute particulate, ions, or residues. They should be low vapor pressure, high auto-ignition, high flashpoint, high lower flammability limit (LFL) materials that are non-toxic and do not contribute chemical species that can coke or poison detritiation systems. Manufacturing solvents and cleaning requirements should be carefully considered during the procurement specification to minimize the impact of tritium introduction to the system (e.g. halogens, hydrocarbons, and particulates).

Tritium compatibility testing is conducted in a variety of ways. SRS has been conducting compatibility testing for many years<sup>56 57</sup>. Stainless steel forgings, welds, and bar stock have been charged with tritium, aged, and tested to determine the effects of hydrogen, helium, and hydrogen and helium combined. In general, these tests are accomplished by preparing samples of the appropriate geometry, acid-cleaning them, placing them in charging vessels and loading them with the desired pressure of tritium at temperatures that do not exceed 350°C. They are then held until they are saturated, quenched to retain the tritium, and aged at -70°C for the required duration to develop the target amount of helium, and then tested.

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<sup>56</sup> Morgan, Michael J. SRNL. *Tritium Aging Effects on the Fracture Toughness Properties of Stainless Steel Base Metal and Welds*. SRNL-STI-2009-00475.

<sup>57</sup> Morgan, M.J., M.H. Tosten, and G.K. Chapman, SRNL. *The Effects of Hydrogen, Tritium, and Heat Treatment on the Deformation and Fracture Toughness Properties of Stainless Steel*, SRNL-STI-2013-00534.

Polymer samples are tested in much the same manner. Samples are loaded into suitable pressure vessels and charged with tritium for the desired duration. The gases generated and mechanical properties are determined. Experiments may be staged with multiple samples to obtain the desired properties as a function of time or dose as needed to support the program. SRNL has extensively used dynamic mechanical analysis to determine the effects of radiation and tritium exposure on the loss modulus of polymers<sup>58 59 60 61 62 63</sup>.

## 5.2 First Wall Design

### 5.2.1 High-Pressure Tritium

For high-pressure tritium, it is generally recommended that the first wall be of all metal construction and hydrogen compatible materials including valves, valve seats, and tubing. The use of non-hydrogen-compatible materials results in material failure and the release of the contained tritium. Elastomers are not tritium-compatible, and, as a result, elastomeric seals and valve seats are not recommended for use in the containment of high-pressure tritium. There are exceptions to this general case, and other criteria may be used when justified by analysis. Additionally, the orientation of high pressure valves with respect to the high pressure tritium is also important. Historically high pressure valves with teflon packed valve stems have been used for long term component storage. This practice was acceptable if the valve was properly oriented where the pressure only wets the metal valve tip. If incorrectly oriented the high pressure tritium is in contact with the teflon packing and the valve will fail.

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<sup>58</sup> Clark, Elliot A. *Radiolytic Gas Production Rates of Polymers Exposed to Tritium Gas*. SRNL, 2013. SRNL-STI-2013-00506.

<sup>59</sup> Clark, Elliot A. *Effects of Gamma Irradiation on EPDM Elastomers*. SRNL, 2013. SRNL-STI-2011-00580, Revision 1.

<sup>60</sup> Clark, Elliot A. and Shanahan, Kirk L. *Effects of Tritium Exposure on UHMU-PE, PTFE, and Vespel* SRNL, 2006. WSRC-STI-2006-00049.

<sup>61</sup> Clark, Elliot A, et al., *Effects of Tritium Gas Exposure on Polymers*. SRNL, 2010. SRNL-STI-2010-00111.

<sup>62</sup> Clark, Elliot A. and Shanahan, Kirk L *Effects of Tritium on UHMW-PE, PTFE, and Vespel Polyimide* SRNL, 2008 WSRC-STI-2008-00281.

<sup>63</sup> Clark, Elliot A. *Tritium Effects on Dynamic Mechanical Properties of Polymeric Materials*. SRNL, 2008. WSRC-STI-2008-00077.

### 5.2.2 Low- and Medium-Pressure Tritium

For the containment of low- and medium-pressure tritium, it is generally recommended that the first containment wall be of all metal construction of hydrogen-compatible materials where possible, including valves, valve seats, and tubing. Hydrogen and helium embrittlement of the materials of construction is not usually significant at low and medium pressures. As a result, non-hydrogen-compatible materials may be used if required by the design or if the required component is not available in other materials.

It is difficult to design a vacuum system, which, in some cases is the first wall, without including some non-hydrogen-compatible materials and elastomers. However, embrittlement of the materials is not an issue because the tritium exposure is transient and the pressure is low. Under these conditions, the elastomers are not exposed to tritium continuously, and most can be used in tritium operations under this condition. Surveillance and/or preventive maintenance schedules should be selected in order to maintain elastomer functionality.

## 5.3 Secondary Wall Design

### 5.3.1 High-Quality Secondary

The design requirements for a high-quality secondary wall are the same as the primary wall. If the secondary wall is required to provide long-term containment of high concentrations of tritium, it should meet the same requirements as the primary or first-wall container.

### 5.3.2 Medium-Quality Secondary

If the secondary wall is a glovebox and only contains tritium that has been diluted by the glovebox gases for a short duration (e.g., a few hours) while the glovebox is cleaned up by the tritium removal system, the requirements can be relaxed. Although the quantity of tritium contained may be quite large, the low pressure and concentration of tritium will not result in material failure due to tritium exposure.

### 5.3.3 Low-Quality Secondary

If the secondary wall is a room or building and only contains tritium that has been diluted by the air contained in the room or building while the room or building is cleaned up by the tritium removal system, the construction requirements can be relaxed. Although the quantity of tritium contained may be quite large, the low pressure and concentration of tritium will not result in material failure.

## 5.4 Cleanup System Design

Most of the components of the tritium removal and cleanup system are only exposed to tritium at low concentrations and pressure for short time periods. The only long-term exposure is in the water collection system where the water is collected at low pressure on a molecular sieve. All-metal construction is recommended, but the materials of construction are not required to be hydrogen-compatible materials. Appropriate, elastomeric sealing materials have been used successfully in these systems for many years without significant problems. When possible, metal seals should be used because they are more durable and reliable and require less maintenance than elastomeric seals.

To minimize the potential for the generation of mixed waste and to decrease radiation exposure of the workers, oil-free pumps should be used where possible.

## 5.5 Storage System Design

Storage systems should consider the total cost of the storage cycle and the purpose for the storage. Storage techniques that increase the complexity of the handling process without adding beneficial features should not be used. The barrier concept discussed in Sections 4.1.1 through 4.1.3, in addition to the wall design considerations discussed above, should be incorporated into all storage system designs.

### 5.5.1 Short-Term Storage

Tritium used to support the day-to-day activities in a facility has to be readily available to the facility customers. If the facility uses tritium in gaseous form and its decay to helium does not impact the process, then, to simplify the operation and the equipment, the tritium can be stored in gaseous form. The storage container should be fabricated of all metal, hydrogen-compatible materials including valves, valve seats, and seals.

### 5.5.2 Medium-Term Storage

If tritium is only used in periods of two years or less, the requirements do not change significantly from those of short-term storage. Experience has shown that tritium can be stored safely at near atmospheric pressure for long periods of time. If the buildup of helium in the supply does not impact the use, then storage as a gas is an acceptable alternative. There are, however, advantages of tritide bed storage for medium-term use. Impurities such as nitrogen and oxygen form uranium nitride and uranium oxide and are removed from the gas stream as the bed is heated and cooled. Helium, which accumulates due to the decay of tritium, and any other impurities remain in the overpressure gas above the bed and may be pumped off after the bed had been cooled down and

the tritium has been gettered by the uranium. As a result, the uranium bed not only provides for tritium storage but also provides a means of maintaining a reasonably pure and stable tritium supply.

### 5.5.3 Long-Term Storage

Due to its short half-life, storing tritium for several years implies that it is not readily needed. It should be placed in a safe and stable condition while it decays.

#### 5.5.3.a Storage as a Gas

Metal tritides have the advantage of significantly decreasing the volume required to store tritium without increasing the pressure of the gas during storage. Compared to the fabrication and preparation of metallic storage beds, regardless of what metal is used, the cost of storage of tritium as a gas at near atmospheric pressure is economical. ASME code-designed stainless steel tanks are available or can be designed and fabricated at a reasonable cost. A tank at atmospheric pressure would have a final pressure of 15 pounds per square inch after all of the tritium had decayed, so embrittlement is not an issue. The long-term storage of hydrogen and tritium in containers is well understood in comparison to the understanding of long-term storage of metal tritides.

#### 5.5.3.b Storage as a Metal Tritide

Uranium beds designed at Sandia in the late 1970s for laboratory use were about the size of two one-gallon paint cans. This included the secondary containment system and electric heaters used to drive off the tritium during removal. These beds were designed to store 50 grams of tritium as uranium tritide that was easily recoverable in a matter of less than an hour. Long-term storage in this type of container is expensive, but the tritium can be easily and quickly recovered for use. Additionally, large uranium beds capable of storing 1,000 liters of hydrogen were used at Mound. Also, impurities such as nitrogen and oxygen form uranium nitride and uranium oxide, and are removed from the gas stream as the bed is heated and cooled. Helium, which accumulates due to the decay of tritium, and other non-reacting impurities remain in the overpressure gas above the bed.

To help resolve unknowns regarding consequences of air-ingress accidents in uranium beds, a series of air-ingress experiments was conducted at Ontario Hydro Research Division, with the participation of PPPL and the Idaho National Laboratory. The experiments indicated that the resulting reaction was restrained with only modest temperature excursions. This leads to the conclusion that the hazards associated with an air-ingress accident involving a uranium bed is smaller than previously anticipated. Additionally, tests conducted by WSRC indicate that, except for catastrophic container failure, the tritium release due to air in-leakage into a uranium tritide bed is limited by diffusion.

Titanium hydride is not pyrophoric at room temperatures, is a stable material, and has been studied for use in the long-term storage of tritium. It is reported to be less prone to spontaneous ignition in air than the parent metal. Following the hydrating process, if the titanium hydride is exposed to air under controlled conditions, a small quantity of hydrogen is released from the material as the oxide layer forms on the surface of the material. Following formation of the oxide layer, titanium hydride is stable in air. Hydrogen will not be released unless the material temperature is significantly increased.

Palladium tritide is not pyrophoric at room temperature, is a stable material, and the overpressure of tritium over the bed at room temperature is approximately 50 torr. Metal tritides have the advantage of significantly decreasing the volume required to store tritium without increasing the pressure of the gas during storage.

## 5.6 Surveillance and Maintenance

The level of equipment surveillance (including radiological monitoring) and maintenance required is based on the hazard class of a facility; i.e., Hazard Category 1 through 3 or Radiological. The specific requirements for the different classes of facility equipment are a function of the safety issues associated with the equipment. These are specified in the facility safety analysis report or other facility safety documentation and in the facility maintenance plan.

## 5.7 Seismic Considerations

This section describes 1) DOE NPH requirements for protection against natural phenomena such as earthquakes, and 2) seismic design and evaluation of equipment and distribution systems. Seismic and wind design and evaluation of structures and facilities are discussed in Section 4.4.

### 5.7.1 DOE Natural Phenomena Hazards Requirements

DOE has requirements for the mitigation of natural phenomena (such as earthquakes, extreme winds, and floods) on its facilities. These requirements are in DOE O 420.1C, *Facility Safety*, and DOE-STD-1020-2012, *Natural Phenomena Hazards Analysis and Design Criteria for DOE Facilities*, which is invoked as a requirement by O 420.1C. DOE O 420.1C provides the overall requirements for mitigation of the effects of natural phenomena. DOE-STD-1020 defines the basic performance requirements for SSCs subjected to NPH loads.

Section 2.2 in DOE-STD-1020-2012 describes the process of determining an NPH design category for each SSC based on the radiological consequences of its failure. It references a categorization scheme described in Appendix A of DOE-STD-1189-2008, *Integration of Safety Into the Design Process*, as well as ANSI/ANS-2.26-2004, *Categorization of Nuclear Facility Structures, Systems, and Components For Seismic Design*. An equipment list should be developed to show the NDC for each SSC identified.

Chapters 3-8 of DOE-STD-1020-2012 contain requirements for characterizing various natural hazards (seismic, wind, flooding, lightning, precipitation, and volcanic eruption) and determining appropriate performance goals and design basis loads to withstand these hazards. These chapters rely extensively on national consensus standards for the hazard characterization and design requirements. The rigor of required hazard characterization increases for SSCs categorized as NDC-3 or higher. Site-specific hazard characterization is not required if all SSCs are NDC-2 or lower; hazard characterization and design criteria need only follow ASCE/SEI 7-10, *Minimum Design Loads for Buildings and Other Structures*. However, if valid, site-specific hazard values are available, even if a facility is NDC-2 or below, such values are always preferable for use in design.

### 5.7.2 Seismic Design and Evaluation of Equipment and Distribution Systems

In the event of an earthquake, DOE facilities need to have adequate measures for the protection of the public, workers, environment, and investment. Due to the evolutionary nature of design and operating requirements as well as developments in engineering technology, existing DOE facilities embody a broad spectrum of design features for earthquake resistance. These features depend on factors such as vintage of the facility design and construction and hardware supplier practices at the time of design and construction. The earliest vintage facilities often have the least design consideration for seismic-induced forces and displacements and exhibit the greatest difference between their design basis and current requirements for seismic design criteria for new facilities. Chapter 9 of DOE-STD-1020-2012 discusses periodic reviews of NPH assessments that are mandated by DOE O 420.1C. If a periodic review leads to a new seismic hazard assessment for a site, and the hazard increases, Chapter 9 also describes steps for evaluating seismic design of existing facilities. DOE has developed a Seismic Evaluation Procedure (SEP)<sup>64</sup> that can assist with such evaluations. It provides a technical approach and generic procedures and documentation requirements, which can be used to evaluate the seismic adequacy of existing equipment and distribution systems.

The SEP is intended to provide DOE facility managers, safety professionals, and engineers with a practical procedure for evaluating the seismic adequacy of equipment. Often the approach used to review the seismic capacity of equipment in facilities is to conduct sophisticated evaluations, which can be very time-consuming, complex, and costly. Much of the available funding and time can be spent on analysis rather than on the real objective of increasing the seismic capacity of the equipment. The SEP is designed to be an extremely cost-effective method of enhancing the seismic safety of existing facilities and reducing the potential for major economic loss that can result from equipment damaged by an earthquake.

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<sup>64</sup> DOE/EH-0545, *Seismic Evaluation Procedure for Equipment in DOE Facilities*, March 1997.

The following is a suggested list of topics to consider in the design of new systems and equipment or the evaluation of existing systems and equipment. Representative equipment that may be found in tritium facilities is listed in Table 5-1.

**ELECTRICAL EQUIPMENT**

Batteries on Racks  
 Motor Control Centers  
 Low- and Medium-Voltage Switchgear  
 Distribution Panels  
 Transformers  
 Battery Chargers and Inverters  
 Instrumentation and Control Panels  
 Instruments on Racks  
 Temperature Sensors  
 Computer Data / Storage Systems  
 Alarm Instrumentation  
 Communications Equipment  
 Tritium Monitors

**PIPING AND RACEWAY SYSTEMS**

Cable and Conduit Raceway Systems  
 Piping  
 HVAC Ducts  
 Underground Piping  
 Underground Raceways  
 Stacks  
 Conveyors of Material

**TANKS**

Vertical Tanks  
 Horizontal Tanks and Heat Exchangers  
 Underground Tanks  
 Canisters and Gas Cylinders  
 Miscellaneous Tanks

**MECHANICAL EQUIPMENT**

Fluid-Operated / Air-Operated Valves  
 Motor/Solenoid-Operated Valves  
 Horizontal Pumps  
 Vertical Pumps  
 Chillers  
 Air Compressors  
 Motor-Generators  
 Engine-Generators  
 Air Handlers  
 Fans  
 HEPA Filters  
 Gloveboxes

Miscellaneous Machinery

**ARCHITECTURAL FEATURES**

Unreinforced Masonry (URM) Walls  
 Hollow Clay Tile Walls  
 Suspended Ceilings  
 Raised Floors  
 Storage Racks  
 Cranes  
 Elevators

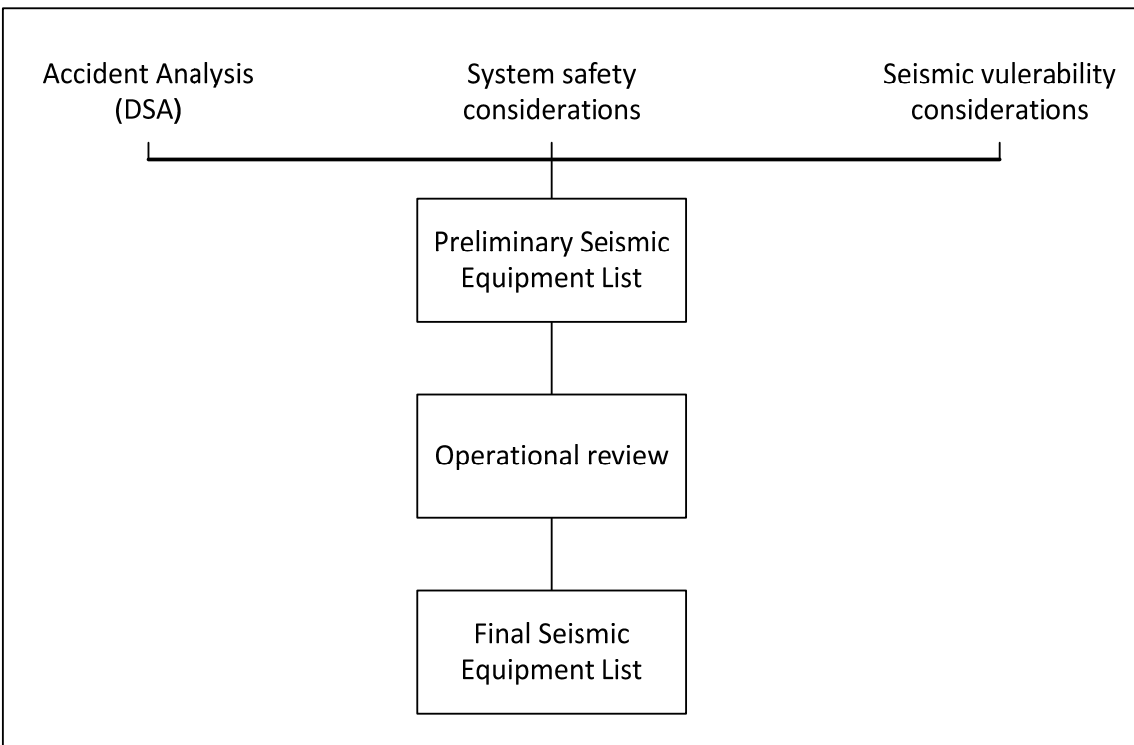
**SWITCHYARD AND SUBSTATION EQUIPMENT**

Power Transformers  
 Miscellaneous Equipment

*4) Table 5-1: Representative equipment found in tritium facilities*

The seismic design category (SDC) for each system or component to be reviewed must be defined (SDC 1, 2, 3, 4, or 5) in the Seismic Equipment List (SEL). The methodology and procedures described in the SEP for evaluating the seismic adequacy of equipment are based on the observed performance, failure, and response of various types of SSCs during and after they were subjected to either actual or simulated earthquake motion. An SSC in a DOE facility can be evaluated for seismic adequacy provided that the associated guidelines, limitations, requirements, and caveats described in the SEP are satisfied.

The general approach for the development of the SEL is envisioned to be a three-step process as depicted in Figure 5-1. After a SEL Team is selected, the first step of the process is the development of the preliminary SEL from a list of the facility SSCs. The SEL Team consists primarily of safety professionals and systems engineers with assistance from seismic engineers and facility operators. Only a portion of the facility SSCs will be contained in the SEL and, in many cases, the SEL will contain only safety-related SSCs that must function during or after a seismic event. The selection of the SSCs belonging on the SEL should be based on the results of accident analyses. These accident analyses should consider the appropriate facility hazards as required by DOE Orders and Standards.



13) Figure 5-1: Development of the seismic equipment list

For the DOE facility being seismically evaluated, accident analyses and their results are typically provided in a DSA. The preliminary SEL should be based on information provided in this DSA. For a nonreactor nuclear facility, DOE-STD-3009-94, Change Notice 3, provides guidance on the preparation of a DSA. Using the guidance in DOE-STD-3009-94, Change Notice 3, and the appropriate accident analyses in the DSA, SSCs can be differentiated into safety-class or safety-significant, and the preliminary SEL can focus on those facility SSCs. For facilities without a DSA, accident analyses comparable to those required for a DSA should be performed.

The preliminary SEL should focus on those SSCs that are classified above as SDC-2 or higher. In addition to selecting SSCs based on an accident analysis or on SDC level, there are system safety considerations and seismic vulnerability considerations that should be addressed when developing the preliminary SEL.

The design or evaluation of SSCs on the SEL should follow the requirements in DOE-STD-1020-2012. SSCs should be assigned an SDC (or NDC for the purpose of broader NPH mitigation) and Limit State per the scheme described in ANSI/ANS-2.26-2004, as modified by Appendix A in DOE-STD-1189-2008. SSCs categorized as SDC-2 or lower should be designed or evaluated using ASCE/SEI 7-10. SSCs categorized as SDC-3 or higher should be designed or evaluated using ASCE/SEI 43-05, *Seismic Design Criteria for Structures, Systems, and Components in Nuclear Facilities*.

Evaluation of existing systems or components must be conducted based on the “Actual” condition of the item. This may be different than the design or “As-Built” condition due to field modifications or deterioration of the item during its service. An examination of the item, its installation, and current condition should be made during a walkdown by seismic engineers as defined in the SEP. Existing systems and components may also be evaluated by use of experience data. This is an alternative approach that can be used if the systems or components are installed in an acceptable manner and meet the rules specified in the SEP to ensure that the item is similar to items in the experience database. DOE has developed references for the evaluation of existing systems and components, has conducted training on their application, and has implemented the seismic evaluation guidelines for systems and components at DOE facilities.

## 5.8 Fire Scenarios

There have been investigations that point to the conclusion that fire scenarios are the dominant risk at most tritium facilities. These fire scenarios are not restricted to seismic-induced fires, but include fires from all sources. These analyses show that the slower burning fires are limiting, and that in these cases, fires result in oxidation rates that are high (in excess of 90 percent). Additionally, compliance with National Fire Protection Association (NFPA) requirements, while limiting the potential for fire spread, does not ensure that a full-facility fire scenario need not be analyzed. LA-UR-02-3803, *Oxidation of Tritium Gas under Accident and Transport Conditions*<sup>65</sup>, describes the oxidation rates that are used in safety analysis at LANL. The bounding value given a fire defaults to 100 percent oxidation for use in safety analyses, as a realistic, less-conservative value has not been able to be identified.

It is possible that fires of sufficient magnitude and frequency may occur at some tritium facilities so as to warrant safety system classifications of both the fire suppression and fire detection systems. This importance that DOE places on fire protection at tritium facilities was highlighted as early as September 1991, during a review of the then-under-construction RTF. The exemption request from some requirements of DOE Orders 5480.7 and 6430.1A for RTF was not approved by HSS/NS until physical modifications were made to the tritium facility<sup>66</sup>. The definition of tritium that was Material at Risk in selected fire (and seismic) scenarios was also developed prior to startup at RTF, which eventually led to decisions on upgraded selected storage containers and inventory limits<sup>67</sup>.

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<sup>65</sup> Jofu Mishima and Chris Steele, “Oxidation of Tritium Gas under Accident and Transport Conditions”, LA-UR-02-3803, July 2002.

<sup>66</sup> DOE Memorandum from S. Blush, NS-1, to R. Claytor, DP-1 and P. Ziemer, EH-1, *RTF Assessment of Fire Barriers*, October 28, 1991.

<sup>67</sup> DOE Memorandum from T. Rollow, Acting Director, Office of Nuclear Safety, to N. Goldenberg, NE-70, *NS Response to Request for Deviation for Replacement Tritium Facility (RTF)*, May 7, 1993.

## 5.9 Instrumentation

This section provides information on several instruments used to detect and monitor tritium. The references are provided for information only. DOE does not certify that a particular product is superior to any other product from another vendor. References in this Standard do not imply endorsement by DOE.

### 5.9.1 Tritium Monitoring Systems

Several different types of instruments may be used to detect and measure tritium in the operation of a facility. Examples and a discussion of such instrumentation follow:

- **Ionization Chambers** — Tritium decays to  $^3\text{He}$  by the ejection of a beta particle. The beta particle generated by the decay of tritium ionizes the surrounding gas. The number of ions produced due to the loss of energy of the beta particle is a function of the type of gas. A sample of gas is collected in the ionization chamber and the ionization current is measured. The resulting chamber ionization current is proportional to the quantity of tritium in the gas. The larger the measuring chambers volume, the higher the output current and the easier it is to measure. However, as the volume of the chamber increases, the longer it will take to get an accurate measurement. Modern electronic systems have solved most of the problems associated with measuring small ionization currents in small volumes and as a result, the volume of the ionization chambers has been reduced over the years from 50 L down to 1 or 2 L. Most tritium measuring instruments have an ionization chamber.
- **Proportional Counters** — Gas proportional counters are also used to measure the amount of tritium contained in a gas. A sample of the gas to be monitored is mixed with a counting gas and passed through a proportional counter tube where the pulses caused by the decay of tritium are counted. Proportional counter monitors can be used for most gas monitoring applications and are also available to measure surface contamination.
- **Scintillation Crystal Detectors** — Scintillation detector systems are used to measure the total mole percent of tritium in a sample of gas independent of the chemical composition of the tritium in the gas ( $\text{HT}$ ,  $\text{DT}$ ,  $\text{T}_2$ , and  $\text{CH}_x\text{T}_y$ ). A sample of the gas is introduced into a measurement chamber at low pressure, generally less than a few torr. The chamber contains a scintillation crystal, which is exposed to the tritium as it decays. The light pulse produced in the scintillation crystal is either counted or is used to produce a current, which is proportional to the mole percent tritium contained in the gas sample. See reference 66 for expanded discussion<sup>68</sup> Crystal

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<sup>68</sup> Ellefson, R.E.; Ellefson Anal. Services, Centerville, OH: Price, B.R. LANL and West, D.S. LANL, *Tritium inventory measurement by beta scintillation detection*, Fusion Engineering 1995, SOFT '95. Seeking a New Energy Era., 16th IEEE/NPSS Symposium (Volume: 2).

scintillation detection is generally used to measure the mole percent of tritium in gases containing high concentrations of tritium.

- **Mass Spectrometer** — Magnetic sector, quadrupole, and drift tube mass spectrometers are used as analytical tools to measure the individual components that make up the gas being measured. Mass spectrometers are generally used for the purposes of assay and accountability or for scientific purposes. A sample of the gas to be measured is introduced at low pressure (a few microns) into a chamber and ionized. The ions produced are then measured by a means that discriminates on mass. The number of ions produced at each mass is measured and is proportional to the partial pressure of the component in the gas sample.

The sophistication of the measurement systems varies greatly from facility to facility throughout DOE. Light isotope, drift tube, mass spectrometers require a large capital investment and require a skilled staff to operate, and, in some cases, may not be cost-effective. All DOE tritium facilities do not require a light isotope drift tube mass spectrometer. Quadrupole mass spectrometers and crystal scintillation detectors are much less expensive, but still require operation by knowledgeable well-trained personnel. The DOE assay and accountability requirements and regulations do not currently reflect this difference in sophistication and cost and currently place the same requirements on small as well as large-scale operations.

- **Liquid Scintillation Counters** — Due to the need to measure the removable tritium on surfaces and in the body water of workers, almost all tritium facilities are equipped with or have access to a liquid scintillation counter. If the scintillation counter is not available on site the service can generally be purchased from a local firm. Liquid scintillation counters are used to measure the quantity of tritium on surfaces, in liquids, and in dissolved samples. For removable surface contamination measurements, a wipe of the surface to be measured is taken using dry paper or a Q-Tip. The filter paper or Q-Tip is then placed in a scintillation cocktail, and the quantity of tritium is measured by counting the light flashes that occur in the scintillation cocktail as the tritium decays. The surface contamination is then calculated in units of dpm/100 cm<sup>2</sup>.

For liquid measurement, a sample of the liquid to be measured is placed in the liquid scintillation cocktail and measured. The tritium concentration of the liquid is calculated in Ci/mL. For solid measurement, a known weight of a material is dissolved to produce a liquid and then the liquid is sampled and measured in the scintillation counter. The quantity of tritium is then calculated in units of Ci/g of the original solid.

- **Raman spectroscopy** has emerged as a reliable method for near real-time analysis of tritium-bearing gas mixtures with potential applications in process control, accountability, and

environmental release calculations. Karlsruhe has reported good results being deployed in a number of applications there<sup>69</sup>.

- **Gas Samplers** — Many different types of gas samplers have been developed and used for measuring very small quantities of tritium in very large volumes of gas. These samplers are used to measure quantities of tritium released through a facility stack and for environmental monitoring at a site. Stack exhaust gas monitoring systems generally use an ionization chamber to measure the tritium in the stack gases and a gas sampler to measure the extremely low levels of tritium that cannot be measured by an ionization chamber.

Most of the stack gas samplers are patterned after the ethylene glycol sampling system developed at Mound Laboratories. A commercial version of this system is now available. In this system, a sample of gas from the stack is circulated through six ethylene glycol bubblers in series. The first three bubblers remove tritium in the form of HTO, DTO, and T<sub>2</sub>O. The gas stream is passed through a heated catalytic reactor where tritium in the form of HT, DT, T<sub>2</sub>, and CH<sub>x</sub>T<sub>y</sub>, is cracked and oxidized to form water. This sample is then passed through three more ethylene glycol bubblers to remove the tritium gas, which is now in the form of water. After a period from a few hours to days, a sample of the ethylene glycol from each bubbler is removed and counted using a scintillation counter to determine the quantity of tritium in each bubbler. Tritium recovered from the first three bubblers is proportional to the tritium in oxide form contained in the stack gases and the tritium recovered in the last three bubblers is proportional to the quantity of tritium in elemental form contained in the exhaust gases.

Due to the extremely small quantity of tritium contained in the atmospheric gases surrounding a tritium facility, the environmental gas samplers use higher flow rate sampler systems than those required for stack monitoring, and, in general, collect the water on molecular sieve traps. The water collected on the molecular sieve traps is then recovered from the trap and the tritium concentration of the gas passing through the trap is calculated from the tritium concentration of the collected water, the gas flow rate through the trap, and the sampling time.

- **Portable Room Air Monitors** — There are several handheld portable room air monitors on the market, and their capabilities and ranges vary as a function of the different manufacturer and the purpose for which they were designed. It is convenient in some activities to have the capability to connect a small hose to the monitor so that it may be used to detect tritium leaks around equipment.
- **Fixed Station Room Air Monitors** — Fixed station monitors are designed to be installed in fixed locations and to be used to monitor the room air tritium concentrations. Depending upon the

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<sup>69</sup> M. Schlösser, *How Can Raman-Inactive Helium be Made Visible in Raman Spectra of Tritium-Helium Gas Mixtures?* Presented at the Tritium 2013 Conference, Nice, France

manufacturer they may have several ranges and are equipped with one or two alarm set points and audible as well as visual alarms.

- **Glovebox Atmosphere Monitors** — Glovebox monitors may be open mesh or closed ionization chambers and are designed to monitor the higher levels of tritium inside the glovebox containment systems.
- **Hood and Exhaust Duct, Air Monitors** — Hood and exhaust duct air monitors are similar to fixed station monitors in range and characteristics.
- **Exhaust Stacks, Air Monitors** — Exhaust stack monitors are similar to fixed station air monitors except that they generally have larger ionization chambers to increase the sensitivity of the monitor.
- **Personnel Friskers and Breath Analyzers** — There has been some interest in instruments which can be used to frisk personnel as they enter and exit tritium contaminated areas. One DOE facility implemented a process of personnel frisking consisting of the use of skin surface wipes counted in a liquid scintillation counter upon entry and exit from tritium contaminated areas. In another facility, a hand station based on counting the associated gas flow across the hands was used. To date, the development work required relating measurements made by these techniques to dose or worker exposure has not been completed. It is expected that differences in the body chemistry of personnel and differences in the time delay between tritium exposure and equilibration of tritium in the body will continue to make the results of skin surface contamination measurement and breathe analysis monitoring inconsistent. The impact of false alarms and inconsistent results on worker confidence will probably continue to make these systems unsatisfactory for worker monitoring.

### 5.9.2 Specialized Instrumentation

There are many other types of specialized devices and/or instrumentation vendors, and some may be superior to those discussed here. No endorsement of these devices should be inferred by the reader.

#### 5.9.2.a Remote Field Tritium Analysis System

A system for the remote, *in-situ* analysis of tritium in surface and ground waters has been developed at SRS. Using automated liquid scintillation counting techniques, the Field Deployable Tritium Analysis System (FDTAS) has been shown to have sufficient sensitivity to measure tritium in water samples at environmental levels (10 becquerels (Bq)/L [ $\sim 270$  pCi/L] for a 100-minute count) on a near-real-time basis.

The FDTAS consists of several major components: a multi-port, fixed-volume sampler, an on-line water purification system using single-use “tritium columns,” a tritium detector employing liquid scintillation counting techniques, and the serial communications devices. The sampling and water purification system, referred to as the “autosampler”, is controlled by a programmable logic controller pre-programmed to perform a well-defined sampling, purification, and flushing protocol. The tritium analyzer contains custom software in the local computer for controlling the mixing of the purified sample with liquid scintillation cocktail, counting, and flushing the cell. An external standard is used to verify system performance and for quench correction. All operations are initiated and monitored at the remote computer through standard telephone line modem communications<sup>70</sup>. This system has been commercially licensed by SRS to a firm that has a line of other sampling monitors.

#### 5.9.2.b Surface Activity Monitor

A surface activity monitor (SAM) for measuring tritium on metal (electrically conducting) and nonmetal (electrically nonconducting) surfaces was developed at Ontario Hydro Technologies (now known as Kinectrics, Inc.).<sup>71 72 73</sup>

The monitor detects tritium on the surface and in the near-surface regions by virtue of primary ionization in air due to the outward electron flux from the contaminated surface. The resulting ion pairs are measured by imposing an electric field between the contaminated surface and a collector plate. A simple theoretical model relates the total tritium concentration on a surface to the measured current.

Experiments benchmarking the application of the surface activity monitor on metal surfaces against independent measurement techniques of aqueous dissolution and thermal desorption show equivalence in the total tritium activities measured. Comparison of surface activity monitor measurements with the dry polystyrene smear protocol has shown that the two methods are complementary. Smearing measures the activity removed by the smear action, which can be used to infer the total activity on the surface. Surface activity monitor measurements determine the total activity on the surface, which can be used to infer removable activity. Ontario Hydro has stated that this device is the only surface monitor for tritium that provides an absolute measurement of the total activity on metallic surfaces.

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<sup>70</sup> K.J. Hofstetter, et al., “Field Deployable Tritium Analysis System for Ground and Surface Water Measurements,” accepted for publication in the *Journal of Radioanalytical and Nuclear Chemistry*.

<sup>71</sup> N.P. Kherani, W.T. Shmayda, “Ionization Surface Activity Monitor for Tritium,” *Fusion Technology* 28(3) (1995) 893

<sup>72</sup> W.T. Shmayda, N.P. Kherani, D. Stodilka, “Evaluation of the Tritium Surface Activity Monitor,” *Proceedings of the Symposium on Fusion Technology*, Lisbon, 1996.

<sup>73</sup> N.P. Kherani, W.T. Shmayda, “Monitor for Measuring the Radioactivity of a Surface,” US and European patents pending.

Experiments demonstrating the application of the surface activity monitor on a variety of non-conducting surfaces have been conducted. Some of the non-conducting surfaces examined include paper, concrete, granite, and wood. It has been determined that the best approach to measuring the tritium on non-conducting surfaces is indirectly; that is, a surface smear is taken and the activity on the smear is done by the SAM. Although this can be done with a liquid scintillation counter by using a SAM, no liquid scintillation cocktail is used, and the measurement is immediate.

Currently, the SAM is commercially available through Tyne Engineering (model 7001), which has measurement ranges of 10-1600 nCi/dm<sup>2</sup>. A summary of the technical specifications of the instrument is given in Table 5-2.

### 5.9.3.c Breathalyzer

A Tritium-in-Breath Monitor was at one time being developed in Canada by Scintrex. It was to be an automatic monitor dedicated towards health physics and radiation biology applications. The Tritium-in-Breath Monitor measured levels of exhaled tritium within 5 minutes of sampling, thus saving considerable time and effort in the monitoring process. This rapid assessment has a sensitivity level of 5 µCi/L-urine equivalent, which may be sufficient for alarming the cautionary levels of in-body tritium. Preliminary development of this equipment had been done at the Atomic Energy of Canada Ltd. research laboratory in Chalk River, Canada. At present, no commercial unit is available.

| TABLE 5-2: Model 7001 Surface Activity Monitor technical specifications |                                                                                                   |
|-------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------|
| Range                                                                   | 0-3552 kDPM/dm <sup>2</sup> (10-1600 nCi/dm <sup>2</sup> )                                        |
| Detection Sensitivity                                                   | +/-2.22 KDPM/dm <sup>2</sup> (+/- 1nCi/dm <sup>2</sup> )                                          |
| Response Time                                                           | <2 minutes                                                                                        |
| Measurement Area                                                        | 100 cm <sup>2</sup>                                                                               |
| Display                                                                 | 128x64 Graphic LCD Display                                                                        |
| Accuracy                                                                | +/- 4.44 kDPM/dm <sup>2</sup> (0-100 kDPM/dm <sup>2</sup> ), +/-10% above 100kDPM/dm <sup>2</sup> |
| Process/Ambient Temperature                                             | 15°C to 50°C                                                                                      |
| Relative Humidity                                                       | 5 to 65%                                                                                          |
| Air Pressure                                                            | 80-120kPa                                                                                         |
| Tritium Wetted Parts                                                    | 316L Stainless Steel, Teflon                                                                      |
| Dimensions                                                              | 150 mm dia x 178 mm high x 254 mm wide                                                            |
| Weight                                                                  | 2.7 kg                                                                                            |

5) Table 5-2: Model 7001 Surface Activity Monitor technical specifications

## APPENDIX A: USEFUL NUMERICAL VALUES

### A.1 General Data

- 1 becquerel (Bq) = 1 disintegration/second =  $2.7 \times 10^{-11}$  Ci
- 1 Ci =  $3.7 \times 10^{10}$  disintegrations/second =  $3.7 \times 10^{10}$  Bq = 37 GBq
- Avogadro's Number =  $6.023 \times 10^{23}$  molecules/mole
- STP conditions: 760 Torr, and 0°C (1 atm and 273K)
- 1 mole of ideal gas at STP = 22.414 L at 0° C, and approximately 24.2 L/mole at room temperature
- 1 Sievert (Sv) = 83.8 roentgens = 100 rem
- A1 quantity-special form
- A2 quantity –other than special form or normal form

### A.2 General Tritium Data

- Tritium decays to  $^3\text{He}$  + beta + neutrino
- Half-life of tritium (Scientific purposes) =  $12.323 \pm 0.004$  years (4500.88  $\pm$  1.46 days)  
Half-life of tritium (Accountability purposes) = 12.33 +/- 0.06 years (DOE M 474.1-2, Figure IV-2)
- Tritium decay factor = 0.99984601/day
- Maximum beta energy of decay (E max.) = 18.6 keV
- Mean beta energy of decay (E mean) = 5.69 keV
- Volume of 1 Ci of tritium ( $\text{T}_2$ ) at STP = 0.386 mL
- Tritium ( $\text{T}_2$ ) gas = 9619 Ci/g
- $\text{T}_2$  gas contains 58023 Ci/mole
- 2.589 Ci/cm<sup>3</sup> of  $\text{T}_2$  at STP
- Diameter of a tritium atom (approximate) = 1.1 Angstroms
- Dissociation energy,  $\text{T}_2$  to 2T = 4.59 eV
- Ionization energy, T to  $\text{T}^+$  +  $\text{e}^-$  = 13.55 eV
- Atomic weight = 3.01605
- Gram molecular weight of tritium = 6.0321 g
- Grams tritium/liter at STP = 0.269122 g/L
- Liters/gram of tritium at STP = 3.71579 L/g
- Boiling point of tritium at 1 atmosphere = 25.0 K
- $\text{T}_2$  gas, at 1 atmosphere pressure and 25°C = 2.372 Ci/cm<sup>3</sup>
- Tritiated water,  $\text{T}_2\text{O}$  = 3200 Ci/cm<sup>3</sup>

**A.3 Regulatory Quantities**

- Type B Quantity = > 1080 Ci
- Type A Quantity = > 0.11 to <1080 Ci depending on form
- Limited Quantity = < 21.6 Ci of tritium in gaseous form
  - < 1.1 Ci of tritium in solid form
  - < 1 Ci of tritium at a concentration of > 1 Ci/L liquid
  - < 100 Ci at a concentration of > 0.1 to < 1 Ci/L liquid
  - < 1000 Ci at a concentration of < 0.1 Ci/L liquid
- A2 Quantity = 40 TBq (1080Ci)
- Limited Quantity Excepted Package Requirement for tritium
  - Radiation level < 0.005 mSv/hr (0.5 mrem/hr)
  - Quantity of radioactive material < limit in 49 CFR 173.425
  - Package meets general design requirements for radioactive material packaging from 49 CFR 173.410
  - Nonfixed/ removable contamination on the external surface of the package is < limit in 49 CFR 173.443 (0.41 Bq of tritium/cm<sup>2</sup> or 22 disintegrations/min (dpm)/cm<sup>2</sup> for tritium)
  - Outside of inner or outer packaging is marked “Radioactive”
- Low Specific Activity (LSA) A quantity of Class 7 (radioactive) material with limited specific activity
  - LSA-I
    - Earth, concrete, rubble or other debris =  $4 \times 10^5$  TBq of tritium/g (0.001 Ci of tritium/g, 1 Ci of tritium/kg or 2.2 lb/Ci of tritium)
  - LSA-II
    - Tritium-Contaminated Water up to: 0.8 TBq of tritium/L (20.0 Ci/L)
    - Solids and Gases up to 0.004 TBq of tritium/g (0.1 Ci/g, 100 Ci/kg)
    - Liquids  $4 \times 10^4$  TBq of tritium/g (0.01 Ci/g, 10 Ci/kg)
  - LSA-III
    - Solids, consolidated wastes, or activated material that meet 49 CFR 173.468 water leach test; and
    - Uniformly distributed in a collection of solid objects or uniformly distributed in solid compact binding agent; i.e., concrete, bitumen, ceramic, etc.
    - Relatively insoluble or intrinsically contained in relatively insoluble material such that, under loss of packaging and placed in water for seven days, would not exceed 4 TBq of tritium (110 Ci of tritium)
    - The average specific activity of the solid does not exceed 0.08 TBq of tritium/g (2.16 Ci/g)
- SCO (Surface Contaminated Object) Non-radioactive solid objects with Class 7 (radioactive) materials distributed on the surfaces
  - SCO-I
    - Nonfixed contamination on accessible surface averaged over 300 cm<sup>2</sup> is < 4 Bq of tritium/cm<sup>2</sup> ( $10^4$   $\mu$ Ci/cm<sup>2</sup>)

- Fixed contamination on the accessible surface averaged over 300 cm<sup>2</sup> is < 4 x 10<sup>4</sup> Bq of tritium/cm<sup>2</sup> (1 μCi/cm<sup>2</sup>)
- Nonfixed plus fixed contamination on the inaccessible surface averaged over 300 cm<sup>2</sup> is < 8 x 10<sup>5</sup> Bq of tritium/cm<sup>2</sup> (20 μCi/cm<sup>2</sup>)
- SCO-II
  - Solid object on which limit for SCO-I is exceeded and meets the following:
    - Nonfixed contamination of accessible surface averaged over 300 cm<sup>2</sup> is < 400 Bq of tritium/cm<sup>2</sup> (10<sup>-2</sup> μCi/cm<sup>2</sup>).
    - Fixed contamination of accessible surface averaged over 300 cm<sup>2</sup> is < 8 x 10<sup>5</sup> Bq of tritium/cm<sup>2</sup> (20 μCi/cm<sup>2</sup>).
    - Nonfixed plus fixed contamination of accessible surface averaged over 300 cm<sup>2</sup> is < 8 x 10<sup>5</sup> Bq of tritium/cm<sup>2</sup> (20 μCi/cm<sup>2</sup>).
- Type A Packages limited to 40 TBq of tritium (< 1080 Ci) per package
- Type B Packages for quantities > 40 TBq of tritium (> 1080 Ci) per package
- Graded Safeguards Program
  - Category III: Weapons or test components, containing reportable quantities > 50 g T<sub>2</sub> with isotopic fraction T<sub>2</sub> > 20 percent
  - Category IV: All other reportable quantities
- 
- Facility Categories
  - Hazard Category 1: Category A reactors and facilities designated by PSO.
  - Hazard Category 2: > 30 g of tritium
  - Hazard Category 3: > 1.6 but < 30 g of tritium
  - Radiological Facility:< 1.6 g of tritium

#### ***A.4 Tritium Dose and Exposure Data***

- Biological half-life = 8 to 12 days (oxide); biological half-life of tritides is currently being researched
- Derived Air Concentration (DAC)
  - DAC for HTO = 20 μCi/m<sup>3</sup> = 2 x 10<sup>-5</sup> μCi/mL = 7 x 10<sup>5</sup> Bq/m<sup>3</sup>
  - DAC for HT = 200,000 μCi/m<sup>3</sup> = 2 x 10<sup>-1</sup> μCi/mL = 9 x 10<sup>9</sup> Bq/m<sup>3</sup>
  - DACs for STCs –see Appendix E
- Dose Conversion Factor (DCF)
  - DCF = 0.067 mrem/μCi (inhalation)
  - DCF = 0.1 mrem/μCi (inhalation plus 50 percent allowance for skin absorption)
- Annual Limit on Intake (ALI)
  - ALI for HTO = 80,000 μCi
  - An initial exposure equilibrium urine count of 1 μCi/L equates to approximately a 3-mrem dose.
  - A urine count of 50 μCi/ L for a year equates to approximately a 5,000-mrem dose.

- Breathing  $20 \mu\text{Ci}/\text{m}^3$  HTO in air for 8 hours/day, 50 week/year will result in a dose of approximately 5,000 mrem
- Tritiated water is approximately 10,000 times more hazardous than tritium gas because of rapid uptake mechanisms.
- Tritium Beta Particles
  - Range in Air = 4.5 to 6 mm
  - Range in Water = 0.0005 cm
  - Range in Tissue = 0.0007 cm
  - Radiation, 1 mCi in man (70 kg) = 0.0044 rem/day
  - Maximum Penetration =  $0.6 \text{ mg}/\text{cm}^3$

#### **A.5 Tritium Container Data**

- SRS Hydride Transport Vessel (HTV)
  - Reusable container for transporting up to 18 grams of tritium
  - Shipped in UC-609/BTSP
  - Dual port, flow through capable
  - One female, one male port, Cajon® SS-4-VCR
  - Tritium stored as uranium tritide
  - Contains 493 g depleted uranium
  - Stoichiometry Maximum 1:2.9
  - Weight 9.3 lbs.
  - Height 9.995 in
  - Diameter 4.6 in
  - Volume  $690 \text{ cm}^3$
  - Maximum normal operating temperature =  $450^\circ\text{C}$
  - Pressure Limit at Maximum normal operating temperature = 2.9 psia
  - Tritium vapor pressure as a function of temperature at U:T = 1:2.9
  - Dissociation Equations
    - $\text{Log } P_{\text{atm}} = -4038.2/T + 6.074$ ,  $P_{\text{atm}} = 10^{-4038.2/T + 6.074}$
    - $\text{Log } P_{\text{psia}} = -4038.2/T + 7.2413$ ,  $P_{\text{psia}} = 10^{-4038.2/T + 7.2413}$
  - Maximum specified tritium leak rate in std cc =  $<1.26 \times 10^{-7}$  std cc/s.
- SRS Product Vessel (PV)
  - Reusable container for transporting 10 g of tritium in gas form
  - Shipped in UC-609/BTSP
  - Volume 21 L
  - Maximum pressure 1,200 torr
  - Height 30.5 in
  - Diameter 9.875 in
  - Weight 44 lb
  - Single valve, Nupro® SS-4-HS-TW

- Male Nut, Cajon® SS-4-VCR-4
- Female Cap Cajon® SS-4-VCR-CP
- Maximum helium leak rate =  $1 \times 10^{-7}$  STP cc/s He with 8.5 psig helium internal pressure by belljar method
- SRS Hydride Storage Vessel (HSV)
  - Container for storing up to 1600 STP liters of hydrogen isotopes
  - Capable of being shipped in UC-609/BTSP
  - Dual port, flow through capable
  - One female, one male port, Cajon® SS-4-VCR
  - Tritium stored as titanium tritide
  - Contains 4400 g titanium (Ergenics HY-STOR 106)
  - Weight 45 lb
  - Height 16.3 in
  - Diameter 6.6 in
  - Maximum normal operations temperature = 760°C
  - Pressure Range at normal operating temperature = 192 psig to Full Vacuum
  - Pressure Limit at 120°C = 1000 psig
  - Maximum helium leak rate =  $1 \times 10^{-7}$  STP cc/s He with 30 psig helium internal pressure by belljar method
- SRS Bulk Tritium Shipping Package (BTSP)
  - Type B(M) Reusable Shipping Package
  - Authorized Content; Up to 150 grams of Tritium as a gas, solid or adsorbed tritiated water
  - Vapor on molecular sieve materials.
  - Package Gross Weight 650 lb
  - Maximum Content Weight 120 lb
  - Packaging Overpack
  - Height 50.5 in
  - Diameter 24.5 in
  - Containment Vessel (CV)
  - Height 37.5 in
  - Diameter 15 in
  - Available Volume 2,943 in<sup>3</sup>
  - 30.625 in high x 10 in ID
  - CV Weight ~153 lbs
  - CV Design Pressure Limit at 400°F @ 500 psig
- SRS LP-50 (No longer certified for shipping).
  - Container for storing tritium in gas form
  - Volume 50 L
  - Aluminum shell for contamination control and valve protection

- Maximum initial pressure at loading 1,200 torr
- Height 28.3 in
- Diameter 13.2 in
- Product Container Weight 32 lb
- Aluminum Shell Weight 43 lb
- Full Package Weight including drum 260 lb
- Single valve, Hoke Model 4213X2 packless valve
- Male Nut, Cajon® SS-4-VCR-4
- Female Cap Cajon® SS-4-VCR-1-BL
- Maximum helium leak rate =  $7 \times 10^{-8}$  STP cc/s He with 22.7 psia helium internal pressure by belljar method
- Mound AL-M1-5
  - Up to 100,000 Ci absorbed on molecular sieve, silica gel or commercial clay
  - Used for storage and shipment of absorbed tritiated water.
  - Cylindrical vessel 6 5/8 in O.D. by 23 7/8 height
  - 316 Stainless Steel
  - Cap which is screwed onto threaded center post of container containing two self-sealing quick disconnect fittings used for connection to tritium monitors to check for tritium leak from container
  - Cap has two functions; physical protection of valves, fittings and pressure transducer at top of container and also secondary containment. The cap is sealed with four O rings; one face seal at the bottom circumference of the cover, another near the top of the center post and two sealing the quick disconnect fittings in their wells at the top of the cap.
  - Organic compounds should be limited to 1% or less of the water content of the container.
  - Halogen compounds are to be avoided

#### **A.6 Other Data**

- Calorimeter Factor
  - 3.0657 +/- 0.009 g of tritium per Watt
  - 0.3240 +/- 0.0009 Watts/g of tritium
- ANSI N14.5-87 Leakage Test
  - $< 1 \times 10^{-7}$  cm<sup>3</sup>/s /He

## APPENDIX B: DEFINITIONS

**Actual Activity** – The total quantity of radioactive material within a particulate; Sometimes referred to as true activity

**Airborne radioactivity area:** Any area where the measured concentration of airborne radioactivity, above natural background, exceeds or is likely to exceed the derived air concentration (DAC) values listed in Appendix A or Appendix C of 10 CFR Part 835, or where an individual present in the area without respiratory protection could receive an intake exceeding 12 DAC-hrs in a week (10 CFR Part 835).

**As low as reasonably achievable (ALARA):** A phrase (acronym) used to describe an approach to radiation protection to control or manage exposures (both individual and collective to the work force and the general public) and releases of radioactive material to the environment as low as social, technical, economic, practical, and public policy considerations permit (DOE 5400.5 Chg 2).

**Below regulatory concern :** A definable amount of low-level waste that can be deregulated with minimal risk to the public (DOE O 435.1 Chg. 1)

**Best available technology for radioactive effluent control (BAT):** The preferred technology for a particular activity, selected from among others after taking into account factors related to technology, economics, public policy, and other parameters. As used in this Order, the BAT is not a specific level of treatment, but is the conclusion of a selection process in which several alternatives are evaluated (DOE O 435.1).

**Biokinetic Model:** A mathematical model that describes in quantitative terms the retention and transport of a material in the body. The biokinetic model used in ICRP 78 is recommended to evaluate intakes of STCs.

**Breathing Zone Air Sampler (BZA):** An air sampler that draws air from the area close enough to the nose so the sample can be considered representative of the air a person breathes (ANSI Z88.2-1992). An example of a breathing zone air sampler is a lapel monitor.

**Buffer zone:** The smallest region beyond the disposal unit that is required as controlled space for monitoring and for taking mitigative measures, as may be required (DOE O 435.1 Chg 1).

**Byproduct material:** (1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing

special nuclear material, and (2) the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content (Atomic Energy Act of 1954, 42 USC 2011). The Energy Policy Act of 2005 revised this definition as follows: The term “byproduct material” means

- (1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing special nuclear material;
- (2) the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content;
- (3) (A) any discrete source of radium-226 that is produced, extracted, or converted after extraction, before, on, or after the date of enactment of this paragraph for use for a commercial, medical, or research activity; or
  - (B) any material that –
    - (i) has been made radioactive by use of a particle accelerator; and
    - (ii) is produced, extracted, or converted after extraction, before, on, or after the date of enactment of this paragraph for use for a commercial, medical, or research activity; and
- (4) any discrete source of naturally occurring radioactive material, other than source material, that –
  - (A) the Commission, in consultation with the Administrator of the Environmental Protection Agency, the Secretary of Energy, the Secretary of Homeland Security, and the head of any other appropriate Federal agency, determines would pose a threat similar to the threat posed by a discrete source of radium-226 to the public health and safety or the common defense and security; and
  - (B) before, on, or after the date of enactment of this paragraph is extracted or converted after extraction for use in commercial, medical, or research activity.

**Certified waste:** Waste that has been confirmed to comply with disposal site waste acceptance criteria (e.g., the Waste Isolation Pilot Plant-Waste Acceptance Criteria for transuranic waste, the DOE/NVO-325 criteria) under an approved certification program (DOE O 435.1 Chg 1)

**Confinement system:** Any equipment, structure, or system, which limits the release and/or dispersion of a hazardous/radioactive material within a facility. Examples are fume hoods, air locks, ventilation systems, and may include containment and recovery systems. Confinement systems may consist of multiple techniques and barriers depending upon the quantity of tritium involved and the consequences of an uncontrolled release (U.S. DOE Tritium Focus Group).

A collection of barriers that can satisfy a specified leak criterion contingent upon operation of its ancillary (active) system. Examples of confinement systems include: a glovebox and its associated cleanup system, and a room with its associated cleanup system. Note that in the context of this definition, a glovebox with an associated glovebox cleanup system is a confinement system. A glovebox structure itself is a containment system if, and only if, the specified leak criterion can be met by the structure itself (DOE-HDBK-1129-99).

An area having structures or systems from which releases of hazardous materials are controlled. Primary confinement systems are process enclosures (gloveboxes, conveyors, transfer boxes, and other spaces normally containing hazardous materials). Secondary confinement areas surround one or more primary confinement systems (operating area compartments) (DOE O 435.1 Chg 1).

**Containment system:** Any equipment, structure, or systems that serve as an integral and essentially leak tight barrier against the uncontrolled release of hazardous/radioactive material to the environment and other areas within the facility. Examples include process piping, sealed containers, tanks, gloveboxes, and any other closed loop system, which holds the material for possible recovery of tritium (U.S. DOE Tritium Focus Group).

A collection of passive barriers that can satisfy a specified leak criterion without operation of any ancillary equipment. An example of a containment system is a series of piping and vessels enclosing tritium gas operations. An example of a simple double containment system is a container within another container with each container acting as a separate and independent containment system; more intricate double containment systems have the capability to monitor the volume between the containers for leak detection of the inner container (DOE-HDBK-1129-99).

The assembly of components of the packaging intended to retain the radioactive material during transport (10 CFR Part 71).

**Controlled Area:** Any area to which access is managed by or for DOE to protect individuals from exposure to radiation and/or radioactive material (10 CFR Part 835).

**Derived Air Concentration (DAC):** For the radionuclides listed in Appendix A of 10 CFR Part 835, the airborne concentration that equals the Annual Limit on Intake (ALI) divided by the volume of air breathed by an average worker for a working year of 2,000 hours, assuming a breathing volume of 2,400 m<sup>3</sup> (10 CFR Part 835).

**Derived Concentration Guide (DCG):** The concentration of a radio nuclide in air or water that, under conditions of continuous exposure for one year by one exposure mode (i.e., ingestion of water, submersion in air, or inhalation), would result in an effective dose equivalent of 100 mrem (1 mSv). DCGs do not consider decay products when the parent radionuclide is the cause of the exposure (DOE Order 5400.5 Chg 2).

**DOE/DOT Type A, approved shipping package:** For the purpose of this text, these are packages that can be used for the transport of Type A quantities of radioactive materials. The two typical packages used for solids are metal 55-gallon drums with full removable lids and metal boxes 4 feet wide by 4 feet high and 7 feet long with removable lids. DOT 7A packages may be fabricated in almost any size to fit special needs like packaging of gloveboxes.

**Documented Safety Analysis (DSA):** Documented safety analysis means a documented analysis of the extent to which a nuclear facility can be operated safely with respect to workers, the public, and the environment, including a description of the conditions, safe boundaries, and hazard controls that provide the basis for ensuring safety (10 CFR Part 830). Successor to the Safety Analysis.

**Dose Assessment:** Process of determining radiation dose and uncertainty included in the dose estimate, through the use of exposure scenarios, bioassay results, monitoring data, source term information and pathway analysis.

**Dose Conversion Factor (DCF):** Dose per unit intake.

**Dosimetric Model:** A mathematical model that prescribes how to use the biokinetic and radiation transport models to quantify the dose to specific organs and tissues and how to calculate effective dose. The dosimetric models for STC are described primarily in ICRP publications 67 and 71.

**Facility segmentation:** The concept of independent facility segments should be applied where facility features preclude bringing material together or causing harmful interaction from a common severe phenomenon. Therefore, the standard permits the concept of facility segmentation provided the hazardous material in one segment could not interact with hazardous materials in other segments. For example, independence of HVAC and piping must exist in order to demonstrate independence for facility segmentation purposes. This independence must be demonstrated and places the “burden of proof” on the analyst. Additionally, material contained in DOT Type B shipping containers (with or without overpack) may also be excluded from summation of a facility’s radioactive inventory if the Certificates of Compliance are kept current and the materials stored are authorized by the Certificate. However, Type B containers without overpack should have heat protection provided by the facility’s fire suppression system (DOE-STD-1027-92, Chg 1, Rev. 1).

**Free liquids:** Liquids that readily separate from the solid portion of a waste under ambient temperature and pressure (DOE O 435.1 Chg 1)

**Gastrointestinal (GI) Tract Model:** A mathematical representation of the behavior of radionuclides in the contents of the human gastrointestinal tract.

**Hazard Category 1:** A facility in which the hazard analysis shows the potential for significant off-site consequences. Category A reactors and facilities designated by PSO. Regardless of the quantity of tritium, a facility that handles only tritium is not a Hazard Category 1 facility unless it is designated so by the PSO (DOE-STD-1027-92, Chg 1, Rev. 1).

**Hazard Category 2:** A facility in which the hazard analysis shows the potential for significant on-site consequences. Facilities with the potential for nuclear criticality events or with sufficient quantities of hazardous material and energy, which would require on-site emergency planning activities. The

threshold tritium inventory for a tritium facility to be designated as a Hazard Category 2 facility is 30 grams or 300,000 Ci (DOE-STD-1027-92, Chg 1, Rev. 1)

**Hazard Category 3:** A facility in which the hazard analysis shows the potential for only significant localized consequences. Facilities included are those with quantities of hazardous radioactive materials that meet or exceed values in Table A.1 of DOE-STD-1027-97. The threshold for tritium is specified as 1.6 grams or 16,000 Ci (DOE-STD-1027-92, Chg 1, Rev. 1)

**Hazardous materials:** Those materials that are toxic, explosive, flammable, corrosive, or otherwise physically or biologically health-threatening (DOE-STD-3009)

**Hazardous wastes:** Those wastes that are designated hazardous by EPA regulations (40 CFR Part 261) (DOE O 435.1 Chg 1) The DOT Hazardous Material Regulations define hazardous materials and hazardous waste differently; see 49 CFR 171.8.

**Insoluble Metal Tritide (IMT):** A type of insoluble special tritium compound in which tritium has formed a chemical bond to a metal

**Insoluble Special Tritium Compound:** A special tritium compound, for which the tritium cannot be rapidly taken up by the systemic compartment of the body

**Insoluble Tritiated Particle (ITP) –** Any tritiated particle from which the tritium is not readily released in air or aqueous solutions during the time interval over which the sample is collected and initially analyzed. This time interval may vary significantly, typically ranging from minutes to days.

**Leak-tight:** A leakage rate, which, “in a practical sense, precludes any significant release of radioactive materials. This degree of containment is achieved by demonstration of a leakage rate that is  $\leq$  to  $1 \times 10^{-7}$  ref  $\text{cm}^3/\text{s}$  of air, at an upstream pressure of 1 atmosphere (atm) absolute, and a downstream pressure of 0.01 atm abs, or less. Note: A leakage rate of  $1 \times 10^{-7}$  ref  $\text{cm}^3/\text{s}$  is equal to  $4.09\text{E-}12$  gram-moles of dry air or helium, and is equivalent to a helium leakage rate, under the same conditions, of approximately  $2 \times 10^{-7}$   $\text{cm}^3/\text{s}$ .” (ANSI N14.5-2014))

**Less than Hazard Category 3 Nuclear Facilities** (replaces the term Radiological Facilities):

Facilities that do not meet or exceed Category 3 threshold criteria specified in DOE STD-1027-92, Rev. 1, Table A.1, but still possess some amount of radioactive material are still Nuclear Facilities. The Category 3 threshold quantity of tritium is 16,000 curies. Less than Hazard Category 3 Nuclear Facilities are exempt from some requirements such as development and maintenance of DSAs but are not exempt from other safety requirements (e.g., 10 CFR Part 820 Subpart A, 10 CFR Part 835). Less than Hazard Category 3 Nuclear Facilities are those with an inventory of radiological materials below the levels as defined in DOE-STD-1027-92, Chg. 1, (currently 16,000 curies). There is no official or legal lower threshold limit to be become a non-nuclear facility for the Less than Hazard Category 3 nuclear facilities.

**Low-level radioactive waste or Low-level waste:** radioactive material that is not high-level radioactive waste, spent nuclear fuel, or AEA section 11.e.2 byproduct material. The phrase is defined in the (Low-Level Radioactive Waste Policy Amendments Act of 1985 and . DOE M 435.1-1 )

**Mixed waste:** Waste containing both radioactive and hazardous waste components as defined by the Atomic Energy Act and the Resource Conservation and Recovery Act, respectively (DOE O 435.1-1 )

**Nonreactor nuclear facility:** those facilities, activities or operations that involve, or will involve, radioactive and/or fissionable materials in such form and quantity that a nuclear or a nuclear explosive hazard potentially exists to workers, the public, or the environment, but does not include accelerators and their operations and does not include activities involving only incidental use and generation of radioactive materials or radiation such as check and calibration sources, use of radioactive sources in research and experimental and analytical laboratory activities, electron microscopes, and X-ray machines (10 CFR Part 830).

**Nuclear facility:** means a reactor or a nonreactor nuclear facility where an activity is conducted for or on behalf of DOE and includes any related area, structure, facility, or activity to the extent necessary to ensure proper implementation of the requirements established by this Part (10 CFR Part 830). DOE-STD-1027-92, Chg. 1 defines tritium inventory thresholds for nuclear facilities.

**Observed Activity:** The apparent quantity of radioactive material within a particulate as determined by liquid scintillation counting, without attempting to correct for beta particle self-absorption, bremsstrahlung, or the emissions from HT or HTO.

**Organically Bound Tritium (OBT):** A type of tritiated material in which the tritium has formed a chemical bond with an organic material – typically via a carbon-tritium bond.

**Primary containment:** The first barrier to an uncontrolled release of hazardous/radioactive material to the environment and/or other areas in the facility. The barrier may/may not serve for containment of the radioactive material (U.S. DOE Tritium Focus Group).

**Protective Action Guides (PAGs):** Projected numerical dose values established by EPA, DOE, or States for individuals in the population. These values may trigger protective actions that would reduce or avoid the projected dose.

**Radioactive waste:** Solid, liquid, or gaseous material that contains radionuclides regulated under the Atomic Energy Act of 1954, as amended, and of negligible economic value considering costs of recovery (DOE O 435.1 Chg. 1).

**Radiological area:** Any area within a controlled area which must be posted as a “radiation area,” “high radiation area,” “very high radiation area,” “contamination area,” “high contamination area,” or “airborne radioactivity area” in accordance with (10 CFR 835.603)

**Receiving and/or shipping area:** An area or two different areas that have been designated as radioactive materials receiving and/or Shipping Areas and have been posted for the receipt and shipment of packaged radioactive materials.

**Receiving and/or Shipping, Storage Area:** An area or two different areas that have been designated as the Receiving and/or Shipping Area, Storage Area and have been posted for the storage of packaged incoming and outgoing shipments of radioactive materials.

**Reference man:** A hypothetical aggregation of human (male and female) physical and physiological characteristics arrived at by international consensus (ICRP Publication 23). These characteristics may be used by researchers and public health workers to standardize results of experiments and to relate biological insult from ionizing radiation to a common base. The “reference man” is assumed to inhale 8,400 cubic meters of air in a year and to ingest 950 liters of water in a year (International Commission on Radiological Protection (ICRP) Publication 89, "Basic Anatomical and Physiological Data for Use in Radiological Protection: Reference Values," September 2001).

**Release of property:** As used in DOE Order 5400.5 Chg. 2, it is the exercising of DOE’s authority to release property from its control after confirming that residual radioactive material (over which DOE has authority) on the property has been determined to meet the guidelines for residual radioactive material in Chapter IV of DOE Order 5400.5 Chg. 2 or any other applicable radiological requirements. There may be instances in which DOE or other authority will impose restrictions on the management and/or use of the property if the residual radioactive material guidelines of Chapter IV of DOE Order 5400.5 Chg. 2 are not met or if other applicable Federal, state, or local requirements cause the imposition of such restrictions (DOE Order 5400.5 Chg. 2).

**Safety analysis:** A documented process that: Provides systematic identification of hazards within a given DOE operation, describes and analyzes the adequacy of the measures taken to eliminate, control, or mitigate identified hazards, and analyzes and evaluates potential accidents and their associated risks (DOE Standard 3009).

**Safety-class structures, systems, and components (safety-class SSCs):** the structures, systems, or components, including portions of process systems, whose preventive or mitigative function is necessary to limit radioactive hazardous material exposure to the public, as determined from safety analyses (10 CFR Part 830).

**Safety-significant structures, systems, and components (safety-significant SSCs):** the structures, systems, and components which are not designated as safety-class structures, systems, and

components, but whose preventive or mitigative function is a major contributor to defense in depth and/or worker safety as determined from safety analyses (10 CFR Part 830).

**Secondary containment:** The second barrier to an uncontrolled release of hazardous or radioactive materials to the environment and/or other areas in the facility. The barrier may/may not serve for containment of the radioactive material (U.S. DOE Tritium Focus Group).

**Self-Absorption Factor for Beta Particles (SAF $\beta$ ):** The fraction of beta particles emitted from within a particulate that escapes the particulate.

**Self-Absorption Factor for Energy (SAFe):** The fraction of energy emitted from within a particulate that escapes the particulate.

**Soil column:** An in-situ volume of soil through which liquid waste streams percolate from ponds, cribs, trenches, drain fields, or other areas or facilities used for the primary purpose of removing or retaining the suspended or dissolved radionuclides contained within the liquid process waste stream (DOE O 435.1).

**Soluble Tritiated Particle:** Any tritiated particle from which the tritium is readily released in air or aqueous solutions during the time interval over which the sample is collected and initially analyzed. This time interval may vary significantly, typically ranging from minutes to days.

**Source material:** (1) uranium, thorium, or any other material which is determined by the commission pursuant to the provisions of section 61 (42 USC 2091) to be source material; or (2) ores containing one or more of the foregoing materials in such concentration as the Commission now DOE and NRC may by regulation determine from time to time (Atomic Energy Act of 1954, 42 USC 2011 et seq.).

**Special nuclear material:** (1) plutonium, uranium enriched in the isotope 233 or in the isotope 235, and any other material which the Commission, pursuant to the provisions of section 51 (42 USC 2071) determines to be special nuclear material, but does not include source material; or (2) any material artificially enriched by any of the foregoing, but does not include source material (Atomic Energy Act of 1954, 42 USC 2011 et seq.).

**Special Tritium Compound (STC):** means any compound, except for H<sub>2</sub>O, that contains tritium, either intentionally (e.g., by synthesis) or inadvertently (e.g., by contamination mechanisms) (10 CFR Part 835).

**Stochastic effects:** means malignant and hereditary diseases for which the probability of an effect occurring, rather than its severity, is regarded as a function of dose without a threshold, for radiation protection purposes (10 CFR Part 835).

**Technical safety requirements (TSRs):** the limits, controls, and related actions that establish the specific parameters and requisite actions for the safe operation of a nuclear facility and include, as appropriate for the work and the hazards identified in the documented safety analysis for the facility: Safety limits, operating limits, surveillance requirements, administrative and management controls, use and application provisions, and design features, as well as a bases appendix (10 CFR Part 830).

**Tritiated Material:** Any material containing at least an accountable amount of tritium

**Tertiary containment:** The third barrier to an uncontrolled release of hazardous or radioactive materials to the environment and/or other areas in the facility. The barrier may/may not serve for containment of the radioactive material (U.S. DOE Tritium Focus Group).

**Treatment:** Any method, technique, or process designed to change the physical or chemical character of waste to render it less hazardous, safer to transport, store or dispose of, or reduced in volume (DOE O 435.1 Chg. 1).

**Uptake:** For STCs, the process by which the tritium atoms in STCs are taken into the systemic compartment of the body. This process includes the uptake of tritium that has been dissociated from the host molecule as well as the uptake of an entire STC molecule

**Waste container:** A receptacle for waste, including any liner or shielding material that is intended to accompany the waste in disposal (DOE O 435.1 Chg 1)

**Waste package:** The waste, waste container, and any absorbent that are intended for disposal as a unit. In the case of surface contaminated, damaged, leaking, or breached waste packages, any overpack shall be considered the waste container, and the original container shall be considered part of the waste (DOE O 435.1 Chg. 1).

## APPENDIX C: ASSAY METHODS

There are a number of different assay methods used at the DOE tritium facilities. Most facilities need assay equipment capable of measuring tritium in gaseous, solid, and liquid form.

- **Gas Analysis:** For assay of gaseous tritium, most facilities use some form of mass spectrometer ranging from quadrupole to large sophisticated light isotope drift tube systems. Gas analysis equipment; especially gas analysis equipment which will measure the low molecular weight gases like  $H_2$ ,  $HD$ ,  $HT$ ,  $D_2$ ,  $DT$  and  $T_2$  accurately; is very expensive and requires a high degree of expertise to operate. These systems can cost from fifty to several hundred thousand dollars.
- **Solid (Metal Tritide):** Tritium stored in solid form such as a metal tritide must either be decomposed to return it to the gas form for analysis, or the heat output of the solid caused by the decay of tritium can be measured in a constant heat flow calorimeter. In order to use calorimetry to measure the tritium quantity, it must be known that the item being assayed does not contain any other radioactive component and that no chemical reactions are taking place in the container. Constant heat flow calorimeters vary in chamber size from a few cubic centimeters up to a few liters, and the item to be assayed must be small enough to fit inside the calorimeter chamber. The constant heat flow assay process is the most accurate assay method available if the chamber size and item to be assayed are well matched.
- **Liquid ( $HTO$ ,  $DTO$ ,  $T_2O$ ):** Tritium at high concentrations in liquid form is generally measured using calorimetry. Low concentrations of tritium in liquid form are generally measured by using a scintillation counter. A sample of the liquid is mixed in a scintillation cocktail, and the quantity of tritium in the sample is measured.

### C.1 Measurement Accuracy and Safeguards and Security

Past DOE directives required that tritium be accounted for to the hundredth of a gram and was a problem in that most of the equipment and the techniques used cannot accurately determine the tritium quantity to a hundredth of a gram once the quantity assayed exceeds about one half gram. Sophisticated equipment was justified mostly for special process needs. Even sophisticated equipment does not measure the quantity of tritium accurately to a hundredth of a gram once the quantity exceeds about five grams. The assay technique to be used in an operation or facility should be discussed with DOE safeguards and security to make sure that it will meet the DOE needs for the facility safeguards and security category and the activities performed in the facility. Current requirements for accountability have been raised to the gram level as discussed in Section 3.1.

## C.2 Tritium Assay Analysis by PVT Mass Spectrometer

The most common method of assaying tritium in gaseous form, i.e., T<sub>2</sub>, HT, and DT, mixed with other gases such as Ar, N<sub>2</sub>, O<sub>2</sub>, and <sup>3</sup>He, is referred to as “PVT mass spec.” The total number of moles of gas, n, in a container is calculated using the equation

$$n = PV/zRT$$

where P = pressure in the container in torr

V = volume of the container in liters

z = compressibility factor (See Table C.1)

R = constant = 62.3631 (See below)

T = temperature (K)

In the formula, the container volume (V) is determined ahead of time by measurement using a volume measuring system, or, if no other means is available, it may be calculated from the physical dimension of the container. The gas pressure (P) and temperature (T) are determined by measurement with available instruments at the time of the mass spec sampling. R is a constant, which is a function of the units of pressure and volume used in the equation and is equal to 62.3631 for pressure in torr and volume in liters.

The compressibility factor (z) is a function of gas type, pressure, and temperature and is either determined from a compressibility table for tritium or estimated using a standard equation such as

$$z_{(T_2)} = 1 + [(P_{\text{torr}} \times 0.000832)/1000]$$

This equation is for a temperature of 295 K.

Most operating facilities have established methods for determining the compressibility factor. For those facilities that do not already have these methods established, Table C-1, *Table of Tritium Compressibility Factors*, may be helpful.

As an example, for a container with a volume of 22.414 L at a pressure of 760 torr and a temperature of 273.15 K (Standard Temperature and Pressure (STP) conditions) the number of moles is calculated as follows:

$$\begin{aligned} N &= PV/zRT \\ &= (760 \times 22.414)/(1.000 \times 62.3631 \times 273.15) \\ &= 1.0000 \text{ mole} \end{aligned}$$

Note:  $z_{(T_2)}$  at 273.15 K = 1.000.

The total moles of gas in the container at any time,  $t$ , is the sum of the moles of the individual gases present in the mixture at that time or in equation form:

$$PV/zRT = n_{(\text{Moles Total})} = n_{(\text{T}_2)} + n_{(\text{HT})} + n_{(\text{DT})} + n_{(\text{CT}_4)} + n_{(\text{qTw})} + n_{(\text{He-3})} + n_{(\text{N}_2)} + n_{(\text{O}_2)} + n_{(\text{Ar})} + n_{(\text{etc.})}$$

The  $n_{(\text{qTw})}$  represents a generic tritium-containing component. The “q” in,  $n_{(\text{qTw})}$ , represents any other element which may be present, and the “w” represents the number of tritium atoms in the molecule. The  $n_{(\text{etc.})}$ , represents any generic, non-radioactive, non-tritium component.

From this equation, it follows that

$$PV/zRT = n_{(\text{Moles Total})} = n_{(\text{Total Moles of Tritium Containing Gases})} + n_{(\text{Total Moles of Non-tritium Gases})}$$

where

$$n_{(\text{Total Moles of Tritium Containing Gases})} = n_{(\text{T}_2)} + n_{(\text{HT})} + n_{(\text{DT})} + n_{(\text{CT}_4)} + n_{(\text{qTw})} + \text{etc.}$$

and

$$n_{(\text{Total Moles of Non-Tritium Gases})} = n_{(\text{He-3})} + n_{(\text{N}_2)} + n_{(\text{O}_2)} + n_{(\text{Ar})} + n_{(\text{etc.})} + \text{etc.}$$

The number of moles of tritium in the container is the sum of the number of moles of tritium in each tritium component. The number of moles of tritium in each tritium component is equal to the number of moles of the component multiplied by the moles of tritium per mole of component. The moles of tritium per mole of component, is defined as the ratio of the number of tritium atoms in the component chemical formula to the number of tritium atoms in  $\text{T}_2$ , i.e., 2.

$$n_{(\text{Total Moles of Tritium})} = \underline{2}n_{(\text{T}_2)} + \underline{1}n_{(\text{HT})} + \underline{1}n_{(\text{DT})} + \underline{4}n_{(\text{CT}_4)}/2 + \underline{w} n_{(\text{qTw})} + \text{etc.}$$

TABLE C-1: Table of tritium compressibility factors at 295 K

$$Z_{(T_2)} = 1 + \{[(P_{\text{atm}} \times 760) \times 0.000832]/1000\}$$

| P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P (atm) | z(T <sub>2</sub> ) | P (atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) |
|------------|--------------------|------------|--------------------|---------|--------------------|---------|--------------------|------------|--------------------|------------|--------------------|
| 0.0        | 1.0000             | 3.0        | 1.0019             | 6.0     | 1.0038             | 9.0     | 1.0057             | 12.0       | 1.0076             | 15.0       | 1.0095             |
| 0.1        | 1.0001             | 3.1        | 1.0020             | 6.1     | 1.0039             | 9.1     | 1.0058             | 12.1       | 1.0077             | 15.1       | 1.0095             |
| 0.2        | 1.0001             | 3.2        | 1.0020             | 6.2     | 1.0039             | 9.2     | 1.0058             | 12.2       | 1.0077             | 15.2       | 1.0096             |
| 0.3        | 1.0002             | 3.3        | 1.0021             | 6.3     | 1.0040             | 9.3     | 1.0059             | 12.3       | 1.0078             | 15.3       | 1.0097             |
| 0.4        | 1.0003             | 3.4        | 1.0021             | 6.4     | 1.0040             | 9.4     | 1.0059             | 12.4       | 1.0078             | 15.4       | 1.0097             |
| 0.5        | 1.0003             | 3.5        | 1.0022             | 6.5     | 1.0041             | 9.5     | 1.0060             | 12.5       | 1.0079             | 15.5       | 1.0098             |
| 0.6        | 1.0004             | 3.6        | 1.0023             | 6.6     | 1.0042             | 9.6     | 1.0061             | 12.6       | 1.0080             | 15.6       | 1.0099             |
| 0.7        | 1.0004             | 3.7        | 1.0023             | 6.7     | 1.0042             | 9.7     | 1.0061             | 12.7       | 1.0080             | 15.7       | 1.0099             |
| 0.8        | 1.0005             | 3.8        | 1.0024             | 6.8     | 1.0043             | 9.8     | 1.0062             | 12.8       | 1.0081             | 15.8       | 1.0100             |
| 0.9        | 1.0006             | 3.9        | 1.0025             | 6.9     | 1.0044             | 9.9     | 1.0063             | 12.9       | 1.0082             | 15.9       | 1.0101             |
| 1.0        | 1.0006             | 4.0        | 1.0025             | 7.0     | 1.0044             | 10.0    | 1.0063             | 13.0       | 1.0082             | 16.0       | 1.0101             |
| 1.1        | 1.0007             | 4.1        | 1.0026             | 7.1     | 1.0045             | 10.1    | 1.0064             | 13.1       | 1.0083             | 16.1       | 1.0102             |
| 1.2        | 1.0008             | 4.2        | 1.0027             | 7.2     | 1.0046             | 10.2    | 1.0064             | 13.2       | 1.0083             | 16.2       | 1.0102             |
| 0.1        | 1.0001             | 4.3        | 1.0027             | 7.3     | 1.0046             | 10.3    | 1.0065             | 13.3       | 1.0084             | 16.3       | 1.0103             |
| 1.4        | 1.0009             | 4.4        | 1.0028             | 7.4     | 1.0047             | 10.4    | 1.0066             | 13.4       | 1.0085             | 16.4       | 1.0104             |
| 1.5        | 1.0009             | 4.5        | 1.0028             | 7.5     | 1.0047             | 10.5    | 1.0066             | 13.5       | 1.0085             | 16.5       | 1.0104             |
| 1.6        | 1.0010             | 4.6        | 1.0029             | 7.6     | 1.0048             | 10.6    | 1.0067             | 13.6       | 1.0086             | 16.6       | 1.0105             |
| 1.6        | 1.0010             | 4.7        | 1.0030             | 7.7     | 1.0049             | 10.7    | 1.0068             | 13.7       | 1.0087             | 16.7       | 1.0106             |
| 1.8        | 1.0011             | 4.8        | 1.0030             | 7.8     | 1.0049             | 10.8    | 1.0068             | 13.8       | 1.0087             | 16.8       | 1.0106             |
| 1.9        | 1.0012             | 4.9        | 1.0031             | 7.9     | 1.0050             | 10.9    | 1.0069             | 13.9       | 1.0088             | 16.9       | 1.0107             |
| 2.0        | 1.0013             | 5.0        | 1.0032             | 8.0     | 1.0051             | 11.0    | 1.0070             | 14.0       | 1.0089             | 17.0       | 1.0107             |
| 2.1        | 1.0013             | 5.1        | 1.0032             | 8.1     | 1.0051             | 11.1    | 1.0070             | 14.1       | 1.0089             | 17.1       | 1.0108             |
| 2.2        | 1.0014             | 5.2        | 1.0033             | 8.2     | 1.0052             | 11.2    | 1.0071             | 14.2       | 1.0090             | 17.2       | 1.0109             |
| 2.3        | 1.0015             | 5.3        | 1.0034             | 8.3     | 1.0052             | 11.3    | 1.0071             | 14.3       | 1.0090             | 17.3       | 1.0109             |
| 2.4        | 1.0015             | 5.4        | 1.0034             | 8.4     | 1.0053             | 11.4    | 1.0072             | 14.4       | 1.0091             | 17.4       | 1.0110             |
| 2.5        | 1.0016             | 5.5        | 1.0035             | 8.5     | 1.0054             | 11.5    | 1.0073             | 14.5       | 1.0092             | 17.5       | 1.0111             |
| 2.6        | 1.0016             | 5.6        | 1.0035             | 8.6     | 1.0054             | 11.6    | 1.0073             | 14.6       | 1.0092             | 17.6       | 1.0111             |
| 2.7        | 1.0017             | 5.7        | 1.0036             | 8.7     | 1.0055             | 11.7    | 1.0074             | 14.7       | 1.0093             | 17.7       | 1.0112             |
| 2.8        | 1.0018             | 5.8        | 1.0037             | 8.8     | 1.0056             | 11.8    | 1.0075             | 14.8       | 1.0094             | 17.8       | 1.0113             |
| 2.9        | 1.0018             | 5.9        | 1.0037             | 8.9     | 1.0056             | 11.9    | 1.0075             | 14.9       | 1.0094             | 17.9       | 1.0113             |
| 3.0        | 1.0019             | 6.0        | 1.0038             | 9.0     | 1.0057             | 12.0    | 1.0076             | 15.0       | 1.0095             | 18.0       | 1.0114             |

Table C-1: Table of Tritium Compressibility Factors at 295 K (*continued*)

$$Z_{(T_2)} = 1 + \{[(P_{\text{atm}} \times 760) \times 0.000832]/1000\}$$

| P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P<br>(atm) | z(T <sub>2</sub> ) | P (atm) | z(T <sub>2</sub> ) |
|------------|--------------------|------------|--------------------|------------|--------------------|------------|--------------------|------------|--------------------|---------|--------------------|
| 18.0       | 1.0114             | 21.0       | 1.0133             | 24.0       | 1.0152             | 27.0       | 1.0171             | 30.0       | 1.0190             | 33.0    | 1.0209             |
| 18.1       | 1.0114             | 21.1       | 1.0133             | 24.1       | 1.0152             | 27.1       | 1.0171             | 30.1       | 1.0190             | 33.1    | 1.0209             |
| 18.2       | 1.0115             | 21.2       | 1.0134             | 24.2       | 1.0153             | 27.2       | 1.0172             | 30.2       | 1.0191             | 33.2    | 1.0210             |
| 18.3       | 1.0116             | 21.3       | 1.0135             | 24.3       | 1.0154             | 27.3       | 1.0173             | 30.3       | 1.0192             | 33.3    | 1.0211             |
| 18.4       | 1.0116             | 21.4       | 1.0135             | 24.4       | 1.0154             | 27.4       | 1.0173             | 30.4       | 1.0192             | 33.4    | 1.0211             |
| 18.5       | 1.0117             | 21.5       | 1.0136             | 24.5       | 1.0155             | 27.5       | 1.0174             | 30.5       | 1.0193             | 33.5    | 1.0212             |
| 18.6       | 1.0118             | 21.6       | 1.0137             | 24.6       | 1.0156             | 27.6       | 1.0175             | 30.6       | 1.0193             | 33.6    | 1.0212             |
| 18.7       | 1.0118             | 21.7       | 1.0137             | 24.7       | 1.0156             | 27.7       | 1.0175             | 30.7       | 1.0194             | 33.7    | 1.0213             |
| 18.8       | 1.0119             | 21.8       | 1.0138             | 24.8       | 1.0157             | 27.8       | 1.0176             | 30.8       | 1.0195             | 33.8    | 1.0214             |
| 18.9       | 1.0120             | 21.9       | 1.0138             | 24.9       | 1.0157             | 27.9       | 1.0176             | 30.9       | 1.0195             | 33.9    | 1.0214             |
| 19.0       | 1.0120             | 22.0       | 1.0139             | 25.0       | 1.0158             | 28.0       | 1.0177             | 31.0       | 1.0196             | 34.0    | 1.0215             |
| 19.1       | 1.0121             | 22.1       | 1.0140             | 25.1       | 1.0159             | 28.1       | 1.0178             | 31.1       | 1.0197             | 34.1    | 1.0216             |
| 19.2       | 1.0121             | 22.2       | 1.0140             | 25.2       | 1.0159             | 28.2       | 1.0178             | 31.2       | 1.0197             | 34.2    | 1.0216             |
| 19.3       | 1.0122             | 22.3       | 1.0141             | 25.3       | 1.0160             | 28.3       | 1.0179             | 31.3       | 1.0198             | 34.3    | 1.0217             |
| 19.4       | 1.0123             | 22.4       | 1.0142             | 25.4       | 1.0161             | 28.4       | 1.0180             | 31.4       | 1.0199             | 34.4    | 1.0218             |
| 19.5       | 1.0123             | 22.5       | 1.0142             | 25.5       | 1.0161             | 28.5       | 1.0180             | 31.5       | 1.0199             | 34.5    | 1.0218             |
| 19.6       | 1.0124             | 22.6       | 1.0143             | 25.6       | 1.0162             | 28.6       | 1.0181             | 31.6       | 1.0200             | 34.6    | 1.0219             |
| 19.7       | 1.0125             | 22.7       | 1.0144             | 25.7       | 1.0163             | 28.7       | 1.0181             | 31.7       | 1.0200             | 34.7    | 1.0219             |
| 19.8       | 1.0125             | 22.8       | 1.0144             | 25.8       | 1.0163             | 28.8       | 1.0182             | 31.8       | 1.0201             | 34.8    | 1.0220             |
| 19.9       | 1.0126             | 22.9       | 1.0145             | 25.9       | 1.0164             | 28.9       | 1.0183             | 31.9       | 1.0202             | 34.9    | 1.0221             |
| 20.0       | 1.0126             | 23.0       | 1.0145             | 26.0       | 1.0164             | 29.0       | 1.0183             | 32.0       | 1.0202             | 35.0    | 1.0221             |
| 20.1       | 1.0127             | 23.1       | 1.0146             | 26.1       | 1.0165             | 29.1       | 1.0184             | 32.1       | 1.0203             | 35.1    | 1.0222             |
| 20.2       | 1.0128             | 23.2       | 1.0147             | 26.2       | 1.0166             | 29.2       | 1.0185             | 32.2       | 1.0204             | 35.2    | 1.0223             |
| 20.3       | 1.0128             | 23.3       | 1.0147             | 26.3       | 1.0166             | 29.3       | 1.0185             | 32.3       | 1.0204             | 35.3    | 1.0223             |
| 20.4       | 1.0129             | 23.4       | 1.0148             | 26.4       | 1.0167             | 29.4       | 1.0186             | 32.4       | 1.0205             | 35.4    | 1.0224             |
| 20.5       | 1.0130             | 23.5       | 1.0149             | 26.5       | 1.0168             | 29.5       | 1.0187             | 32.5       | 1.0206             | 35.5    | 1.0224             |
| 20.6       | 1.0130             | 23.6       | 1.0149             | 26.6       | 1.0168             | 29.6       | 1.0187             | 32.6       | 1.0206             | 35.6    | 1.0225             |
| 20.7       | 1.0131             | 23.7       | 1.0150             | 26.7       | 1.0169             | 29.7       | 1.0188             | 32.7       | 1.0207             | 35.7    | 1.0226             |
| 20.8       | 1.0132             | 23.8       | 1.0150             | 26.8       | 1.0169             | 29.8       | 1.0188             | 32.8       | 1.0207             | 35.8    | 1.0226             |
| 20.9       | 1.0132             | 23.9       | 1.0151             | 26.9       | 1.0170             | 29.9       | 1.0189             | 32.9       | 1.0208             | 35.9    | 1.0227             |
| 21.0       | 1.0133             | 24.0       | 1.0152             | 27.0       | 1.0171             | 30.0       | 1.0190             | 33.0       | 1.0209             | 36.0    | 1.0228             |

To determine the component in the gas and the number of moles of each component in the gas, a sample of the container gas is analyzed, and the mole percent of each gas is determined. This gas analysis results in a number for each component in the mixture, which represents the mole percent of each gas at the time of the analysis. The mole percent (m%) is calculated by

$$m\%_{(\text{component})} = (\text{Moles of a component} / \text{Moles Total}) \times 100$$

Therefore, the Mole Percent Total (m%<sub>(Total)</sub>) is

$$m\%_{(Total)} = 100 = m\%_{(T_2)} + m\%_{(HT)} + m\%_{(DT)} + m\%_{(CT_4)} + m\%_{(qTw)} + m\%_{(He-3)} + m\%_{(N_2)} + m\%_{(O_2)} + m\%_{(etc.)}$$

The number of moles of each gas component in the container is calculated by

$$n_{(Moles\ of\ Component)} = (m\%_{(component)} / 100) \times n_{(Total\ Moles)}$$

The grams of each component can then be calculated using the formula

$$\text{Grams of Component} = (m\%_{(component)} / 100) \times (n_{(Total\ Moles)}) \times (\text{Gram Molecular Weight of Component})$$

The following is the process used to determine the number of moles and grams of tritium in the HT component in a container.

1. Calculate the total moles of material in the container.
2. Analyze the sample using a mass spectrometer to determine the mole percent of HT.
3. Calculate the number of moles of HT in the container.
4. Calculate the number of grams of HT in the container.
5. Multiply the moles of HT by  $\frac{1}{2}$  to determine the moles of  $T_2$  in the HT component.
6. Multiply the moles of HT by  $\frac{1}{2}$  and by the gram molecular weight of tritium to determine the grams of tritium in the HT component.

In steps 5 and 6, the  $\frac{1}{2}$  is used because it is the ratio of the number of tritium atoms in HT to the number of tritium atoms in  $T_2$  ( $HT/T_2 = 1/2$ ).

The percent tritium in the container is the sum of the mole percent of each tritium component multiplied by the number of moles of tritium per mole of the tritium component. The moles of tritium per mole of component is equal to the ratio of the number of tritium atoms in the component chemical formula to the number of tritium atoms in  $T_2$ ; i.e., 2. This can be expressed as the following formula:

$$m\%_{(Tritium\ Per\ Mole\ Total)} = \underline{2} m\%_{(T_2)} + \underline{1} m\%_{(HT)} + \underline{1} m\%_{(DT)} + \underline{4} m\%_{(CT_4)} + \underline{2w} m\%_{(qTw)} + \text{etc.}$$

The moles of tritium in the container are the sum of the moles of tritium contained in each component in the container. This is calculated using the following equation, which is the ratio of the tritium atoms in the component chemical formula to the tritium atoms in  $T_2$ ; i.e., 2. In the formula  $qT_w/T_2$  this would be  $w/2$ , or in  $HT/T_2$  this would be  $1/2$ .

$$\begin{aligned} \text{Moles of Tritium} = & [(m\%_{(T_2)}/100) \times (T_2/T_2) \times n_{(\text{Moles Total})}] + [(m\%_{(HT)}/100) \times (HT/T_2) \times n_{(\text{Moles Total})}] + \\ & [(m\%_{(DT)}/100) \times (DT/T_2) \times n_{(\text{Moles Total})}] + [(m\%_{(CT_4)}/100) \times (CT_4/T_2) \times n_{(\text{Moles Total})}] \\ & + [(m\%_{(qT_w)}/100) \times (qT_w/T_2) \times n_{(\text{Moles Total})}] + \dots \end{aligned}$$

where  $x/T_2$  is the number of moles of tritium per mole of the tritium component.

Factoring out  $n_{(\text{Moles Total})}/100$  and rearranging, the equation becomes

$$\begin{aligned} (\text{Moles of tritium} \times 100)/(n_{(\text{Moles Total})}) &= (m\%_{(T_2)} + m\%_{(HT)} \times 1/2 + m\%_{(DT)} \times 1/2 + m\%_{(CT_4)} \times 2 + m\%_{(qT_w)} \\ &\quad \times w/2 + \text{etc.}) \\ &= m\%_{(\text{Tritium Per Mole Total})} \end{aligned}$$

Rearranging the equation becomes

$$\text{Moles of tritium} = (n_{(\text{Moles Total})} \times m\%_{(\text{Tritium Per Mole Total})})/100$$

The amount of tritium in grams is obtained by multiplying the moles of tritium obtained in the last equation by the gram molecular weight of tritium (6.0321g).

As an example, the grams of tritium in a shipment of research grade tritium are determined using the following process.

An analysis of a shipment of research grade gaseous tritium shows the following:

| Component       | Percent |
|-----------------|---------|
| T <sub>2</sub>  | 99.704  |
| D <sub>2</sub>  | 0.115   |
| DT              | 0.079   |
| HT              | 0.050   |
| HD              | 0.023   |
| <sup>3</sup> He | 0.016   |
| Ar              | 0.012   |
| N <sub>2</sub>  | 0.010   |
| H <sub>2</sub>  | 0.005   |

The container pressure is 742 mm, temperature is 20°C, and volume is 49.348 liters. This gives a compressibility factor ( $z_{(T_2)}$ ) of 1.0006 and the constant, R, is 62.3631.

Calculating the percent tritium is as follows:

$$\begin{aligned}
 m\%_{(\text{Tritium Per Mole Total})} &= m\%_{(T_2)} + m\%_{(HT)} \times 1/2 + m\%_{(DT)} \times 1/2 \\
 &= 99.704 + (0.050 \times 1/2) + (0.079 \times 1/2) \\
 &= 99.704 + 0.025 + 0.0395 \\
 &= 99.7685 \text{ percent}
 \end{aligned}$$

The number of moles of gas in the container is calculated by

$$\begin{aligned}
 n_{(\text{Moles Total})} &= PV/zRT \\
 &= (742 \times 49.348)/(1.0006 \times 62.3631 \times 293.15) \\
 &= 2.002 \text{ moles total}
 \end{aligned}$$

The amount of tritium in grams is

$$\begin{aligned}
 \text{Grams of tritium} &= (n_{(\text{Moles Total})} \times m\%_{(\text{Tritium Per Mole Total})} \times 6.0321)/100 \\
 &= (2.002 \times 6.0321 \times 99.7685)/100 \\
 &= 12.046 \text{ grams tritium}
 \end{aligned}$$

The determination of the amount of tritium is only for the point in time that the sample was analyzed as a result of the decay of tritium. Over time, the number of moles of the gases containing tritium decreases. The pressure and number of moles of non-tritiated gases increase with time due to the  $^3\text{He}$  produced and the molecules formed by the atoms of other materials released when the tritium decays. Therefore, the mole percent and number of moles of gas in the container and in the sample change with time. Figure C-1 shows an example of the changing moles in a gas mixture.

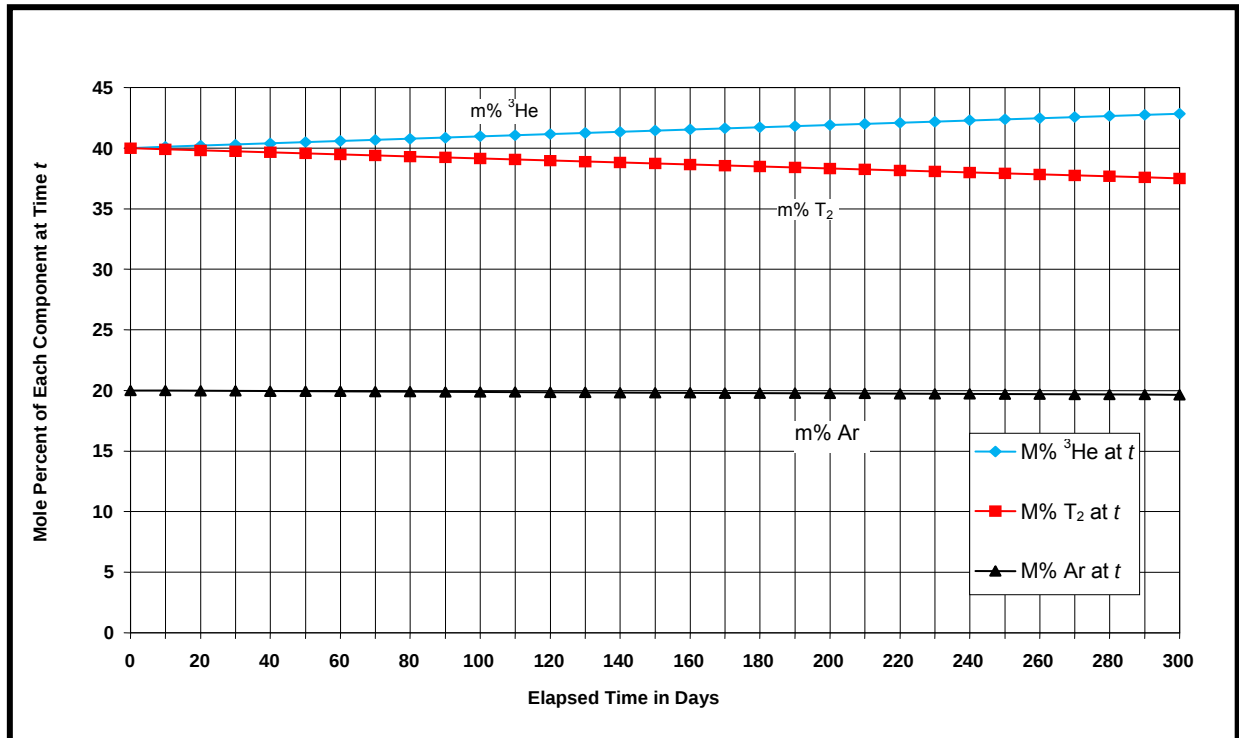


FIGURE C-1. Change in mole percent with time in a mixture of 40%  $\text{T}_2$ , 40%  $^3\text{He}$ , and 20% Ar

The total number of moles in the container is known at the time of the sampling but not at the time of the analysis. If the gas analysis is performed on the same day as the sampling, then, since the half-life of tritium is 4500.88 days, the error caused by the decay is small ( $< 0.015$  percent for 1 day, approximately 1 percent in 67 days). Some facilities are equipped to perform the sample analysis within a few minutes or on the same day as the sampling. Others facilities depend upon collecting a gas sample, on the sampling date, for analysis at a later date.

The following equations include three significant dates. The Sample Date is the day the gas sample for analysis is collected, and the pressure, volume, and temperature of the container are measured and recorded to determine the total moles of gas. The Analysis Date is the date the gas sample is analyzed to determine the mole percent of tritium per mole in the sample. The Book Value Date is a date on which the quantity of tritium in the container is known from a previous assay. Additionally,

it assumes a gaseous mixture at the start of 100% tritium and other stable gases; i.e., it does not contain HT, DT, HTO, or other radioactive or non-inert gases.

The process steps are as follows:

1. Calculating the number of moles of gas in the container on the Sampling Date.
2. Determining the percent of tritium in the collected sample on the Analysis Date by gas analysis.
3. Using the two values in steps 1 and 2, the time period, in days, between the two dates and the half-life of tritium, calculating the number of grams of tritium in the container on the Sampling Date.

Once the quantity of tritium in the container on the Sampling Date is known, the quantity of tritium in the container for any other date, including the Book Value Date can be calculated.

The number of moles of gas in the container on the Sampling Date ( $n_{(TotCSmplDa)}$ ) is the sum of the number of moles of the non-tritiated gases present in the container, which do not change with time due to decay, ( $n_{(Non-tritium)}$ ), plus the number of moles of tritium in the container at the time of the sampling ( $n_{(T2CSmplDa)}$ ). In equation form

$$\begin{aligned} PV/zRT &= n_{(TotCSmplDa)} \\ &= n_{(Non-tritium)} + n_{(T2CSmplDa)} \end{aligned}$$

Rearranging the equation it becomes

$$n_{(Non-tritium)} = n_{(TotCSmplDa)} - n_{(T2CSmplDa)} \quad \{1\}$$

The total number of moles of material in the container on the Analysis Date, ( $n_{(TotCanlDa)}$ ) is

$$n_{(TotCanlDa)} = n_{(Non-tritium)} + n_{(T2CSmplDa)} \times e^{((t \ln(0.5))/4500.88)} + 2 \times (n_{(T2CSmplDa)} - n_{(T2CSmplDa)} e^{((t \ln(0.5))/4500.88)})$$

where  $n_{(Non-tritium)}$  = sum of the moles of the other gases

$(n_{(T2CSmplDa)} (e^{((t \ln(0.5))/4500.88)}))$  = number of moles of  $T_2$  decayed

$2 \times (n_{(T2CSmplDa)} - n_{(T2CSmplDa)} e^{((t \ln(0.5))/4500.88)})$  = number of moles of  $^3He$  created

$t$  = time between the Sampling Date and Analysis Date in days

Factoring, the equation becomes

$$n_{(TotCAnlDa)} = n_{(Non-tritium)} + n_{(T2CSmplDa)} \times (2 - e^{((t \times \ln(0.5))/4500.88)}) \quad \{2\}$$

On the day the gas analysis is performed, the mole percent of tritium ( $m\%_{(T2MAnlDa)}$ ) is determined by gas analysis. The equation is

$m\%_{(T2MAnlDa)}$  is equal to the number of moles of tritium on the sampling date decayed to the analysis date; i.e.,  $(n_{(T2CSmplDa)} \times e^{((t \times \ln(0.5))/4500.88)})$  divided by the number of moles in the sampled container on the analysis date; i.e.,  $(n_{(TotCAnlDa)})$  multiplied by 100 to convert it to percent.

$$m\%_{(T2MAnlDa)} = \{(n_{(T2CSmplDa)} e^{((t \times \ln(0.5))/4500.88)}) / n_{(TotCAnlDa)}\} \times 100$$

where  $(n_{(T2CSmplDa)} \times e^{((t \times \ln(0.5))/4500.88)})$  = number of decayed moles of T  
 $(n_{(TotCAnlDa)})$  = number of moles in the sample on the Analysis Date  
 $t$  = time between the Sampling Date and Analysis Date in days

Rearranging the equation, it becomes

$$n_{(TotCAnlDa)} = \{(n_{(T2CSmplDa)} e^{((t \times \ln(0.5))/4500.88)}) / m\%_{(T2MAnlDa)}\} \times 100 \quad \{3\}$$

Substituting Equation {1} into Equation {2}, we have

$$n_{(TotCAnlDa)} = n_{(TotCSmplDa)} - n_{(T2CSmplDa)} + n_{(T2CSmplDa)} \times (2 - e^{((t \times \ln(0.5))/4500.88)})$$

Rearranging the equation, it becomes

$$n_{(TotCAnlDa)} = n_{(TotCSmplDa)} + n_{(T2CSmplDa)} \times (1 - e^{((t \times \ln(0.5))/4500.88)}) \quad \{4\}$$

Substituting Equation {3} into Equation {4}, we have

$$100 (n_{(T2CSmplDa)} e^{((t \times \ln(0.5))/4500.88)}) / m\%_{(T2MAnlDa)} = n_{(TotCSmplDa)} + n_{(T2CSmplDa)} (1 - e^{((t \times \ln(0.5))/4500.88)})$$

Multiplying through by  $m\%_{(T2MA\text{nlDa})}$ , it becomes

$$100 (n_{(T2CS\text{mplDa})} e^{((t \times \ln(0.5))/4500.88)}) = m\%_{(T2MA\text{nlDa})} n_{(TotCS\text{mplDa})} + m\%_{(T2MA\text{nlDa})} n_{(T2CS\text{mplDa})} (1 - e^{((t \times \ln(0.5))/4500.88)})$$

Rearranging and factoring, we have

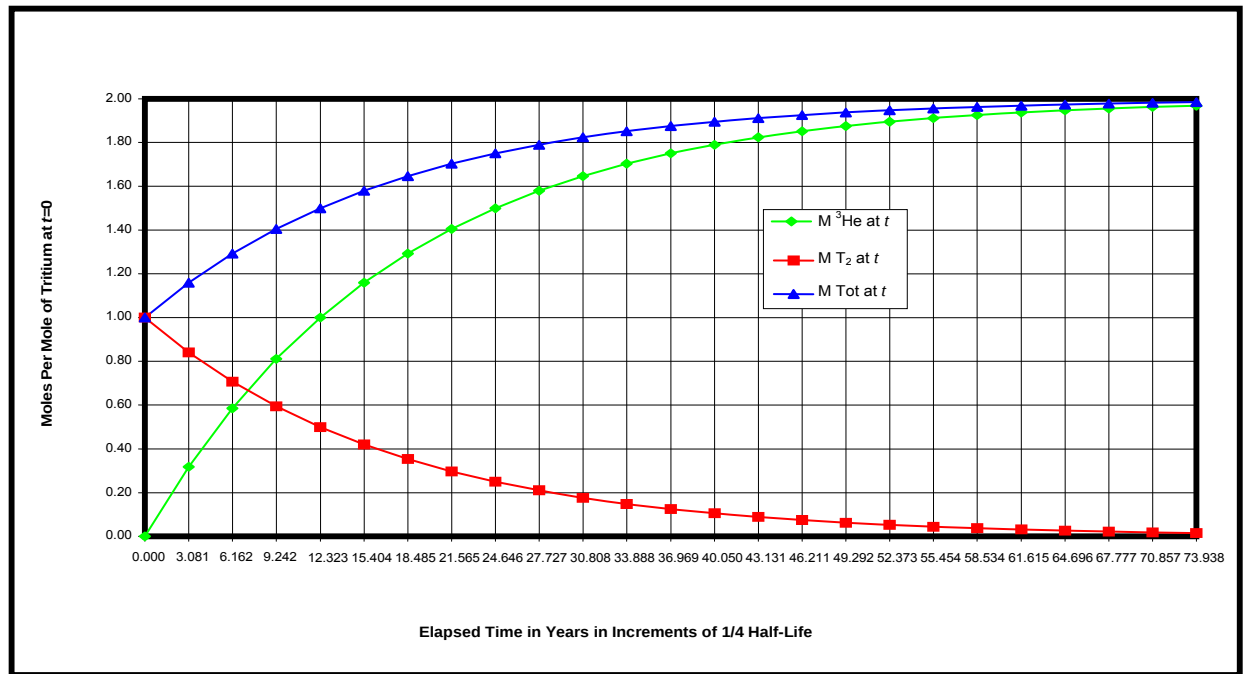
$$n_{(T2CS\text{mplDa})} = (n_{(TotCS\text{mplDa})} \times m\%_{(T2MA\text{nlDa})}) / ((e^{((t \times \ln(0.5))/4500.88)}) \times (100 + m\%_{(T2MA\text{nlDa})})) - m\%_{(T2MA\text{nlDa})})$$

The quantity of tritium in grams is then the number of moles of tritium on the Sampling Date,  $n_{(T2CS\text{mplDa})}$ , multiplied by the gram molecular weight of tritium (6.03210 g) or

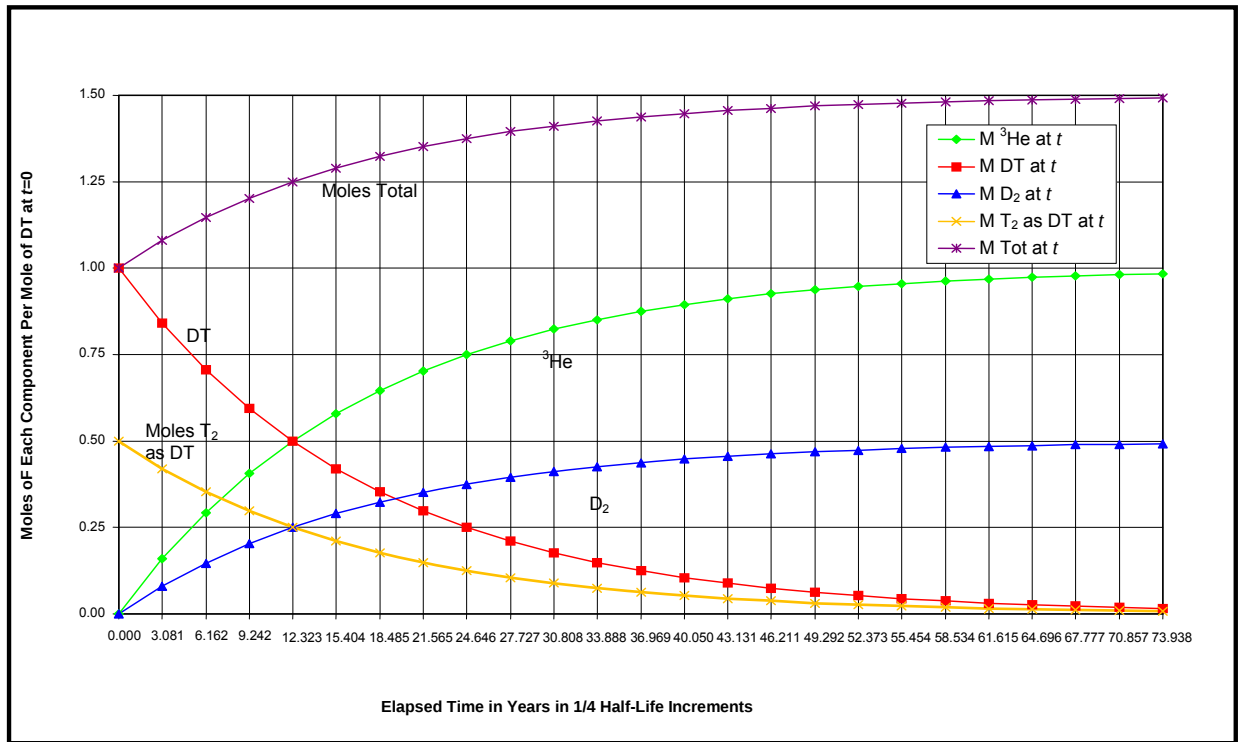
$$\text{Grams } T_2 \text{ On Sampling Date} = \frac{6.0321 \times (PV/zRT) \times (m\%_{(T2MA\text{nlDa})})}{[e^{((t \times \ln(0.5))/4500.88)} \times (100 + m\%_{(T2MA\text{nlDa})})] - m\%_{(T2MA\text{nlDa})}}$$

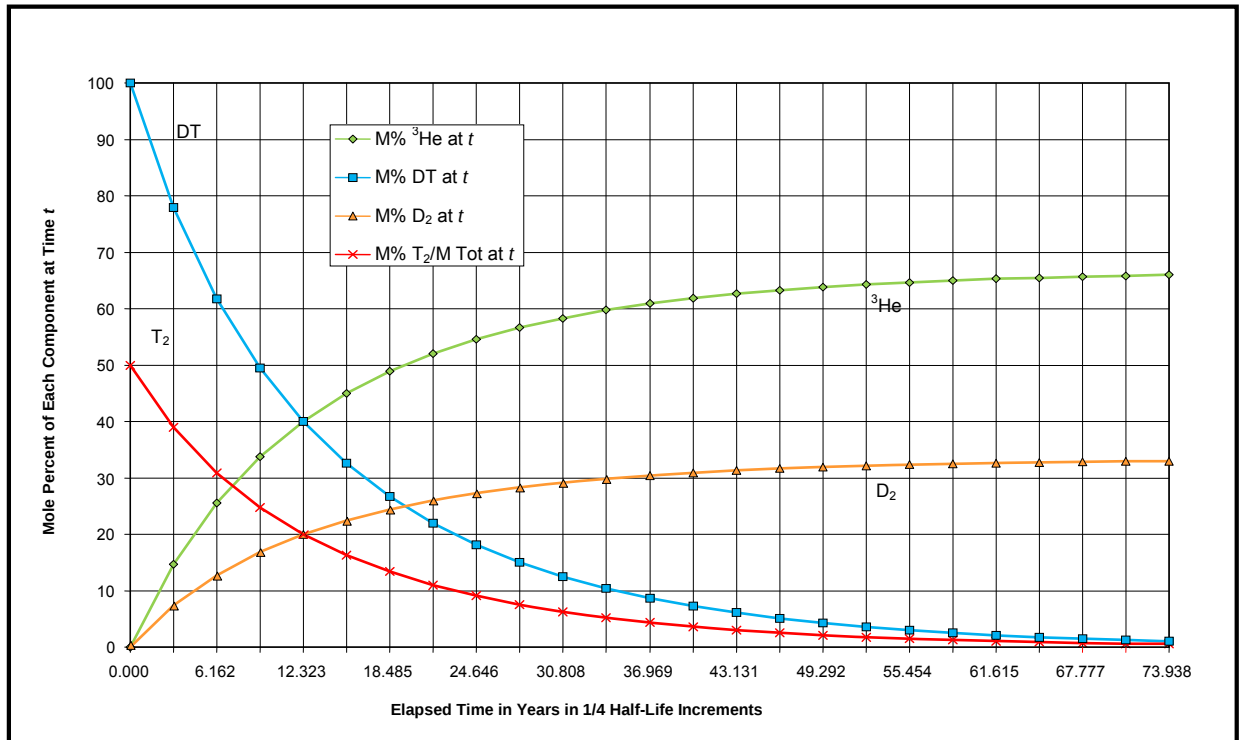
If a container has pure tritium mixed with other non-decaying and non-chemically reacting gases on the Book Value Date, sampled on the Sample Date and analyzed on the Analysis Date, then the gram of tritium in the container on the Sample Date can be calculated using the derived formula, the PV/zRT data and the  $m\%_{(T2MA\text{nlDa})}$  data measured on the Analysis Date.

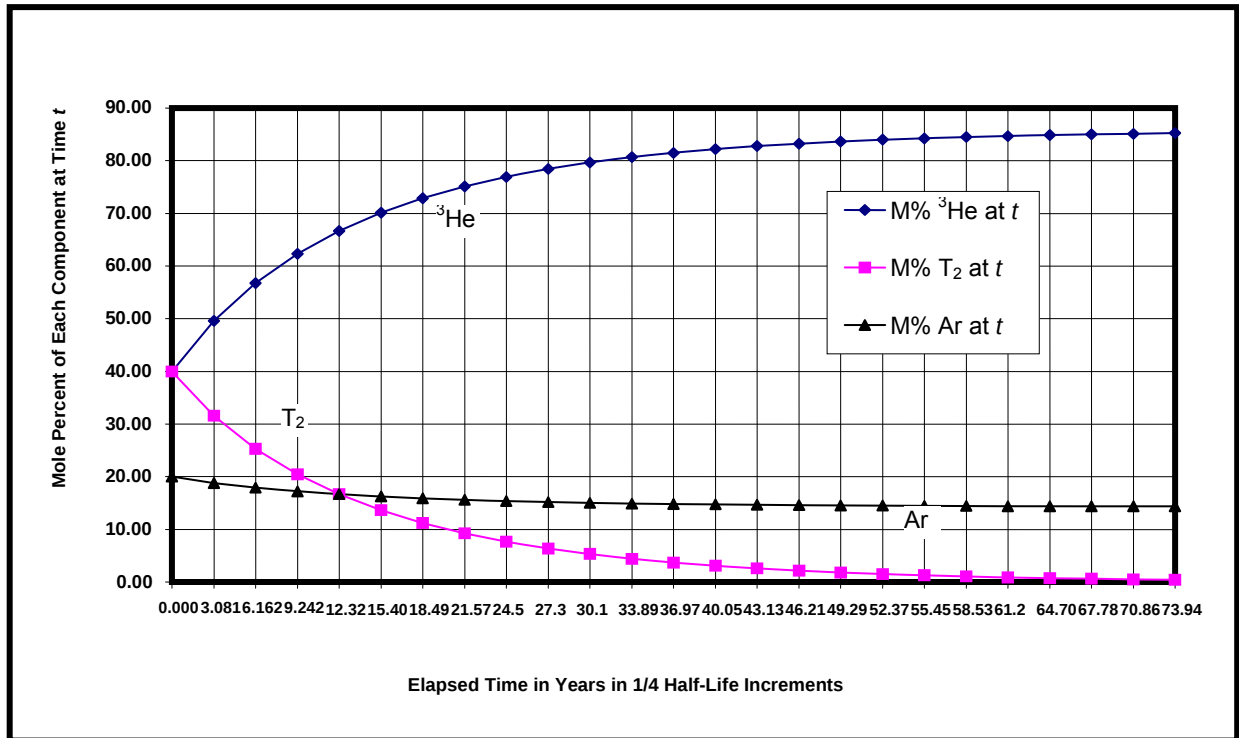
Figure C-2 is a graph of the changes in the moles of material taking place in a container of pure  $T_2$  versus time over a period of six tritium half-lives. The graph shows the moles of tritium decreasing from 1.0 and approaching 0.0, the  $^3\text{He}$  increasing from 0.0 and approaching 2.0, and the total moles increasing from 1.0 and approaching 2.0.

FIGURE C-2. Moles of  $T_2$  and  $^3He$  versus time

Similar formulas can be derived for other two-component gases, such as HT mixed with other non-tritiated gases and DT mixed with other non-tritiated gases. See Figures C-3 to C-6 for other examples.

FIGURE C-3: Moles of DT,  $^3\text{He}$ , and  $\text{D}_2$  versus time

FIGURE C-4: Mole percent of  $\text{T}_2$ ,  $\text{D}_2$ , DT, and  $^3\text{He}$  versus time



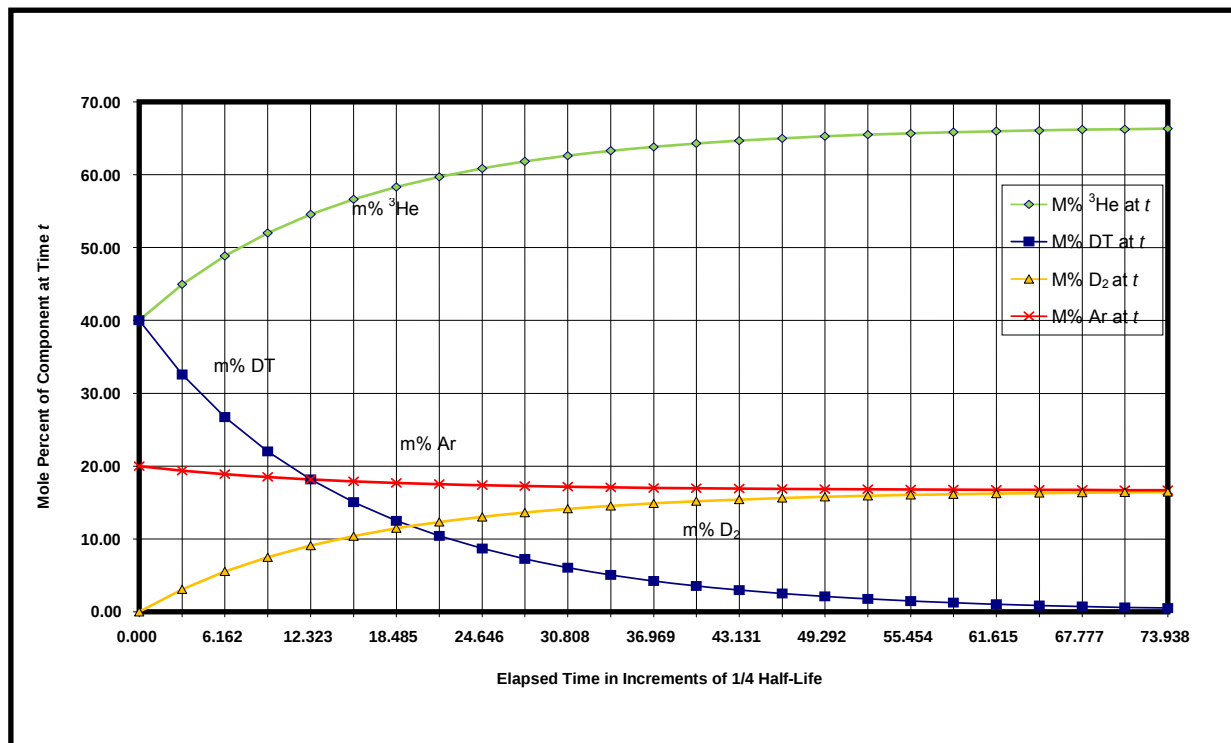


FIGURE C-6: 40% DT, 40%  $^3\text{He}$ , and 20% Ar, Change in mole percent of each component versus time

### C.3 Calorimetry Assay

Calorimetry is the quantitative measurement of heat. A calorimeter is an apparatus for measuring heat quantities generated in or emitted by materials in processes such as chemical reaction, changes of state, and formation of solutions. Heat is generally measured in calories or joules. A calorie is a unit of heat energy equal to the heat energy required to raise the temperature of a gram of water from 14.5 to 15.5°C, at a constant pressure of 1 atmosphere. A calorie is equal to 4.186 joules.

A calorimeter designed to be used in processes that continually generate heat (power sources) and measures power instead of heat is called a Constant Heat Flow (CHF) calorimeter. A CHF calorimeter measures the power (joules/second) of a source not the heat output (joules) of a source. The power is usually measured in Watts, which is a unit of power equal to 1 joule/second.

A radioactive material is a power source, which deposits the energy due to decay in the radioactive material itself and in the materials surrounding the radioactive material. The power generated by the decay of tritium has been measured and is equal to  $0.3240 \pm 0.0009$  watts/gram of tritium.

Mound Laboratory has been the leader in the design, fabrication, calibration, and operation of CHF calorimeters for many years. Mound has specialized in the development of CHF calorimeters to be used in the measurement of radioactive material quantities by measuring their power output. CHF calorimeters are generally designed to meet the specific needs of the items to be assayed and are limited in application by the following:

- Physical size of the calorimeter measurement chamber,
- Wattage range of the measurement system,
- Precision and accuracy of the measurement for the size and wattage range of the item to be measured,
- Throughput or number of samples to be measured per day.

CHF calorimeters have been designed in many different configurations, such as over/under, and twin. Most CHF systems in use today use digital control systems operated by a stored program and are easy to operate. The steps in making a CHF measurement are generally as follows:

- Install a dummy mass in the calorimeter container, pack steel or copper wool around the dummy mass, and install it in the measurement chamber.
- Make a zero baseline run at a wattage level ( $W_{zbl}$ ), which is at a wattage level greater than the unknown wattage level of the sample to be measured.
- During the baseline run, the digital control system establishes a calorimeter bridge voltage value for a known ( $W_{zbl}$ ) wattage input.
- Remove the calorimeter container from the measurement chamber, remove the dummy sample from the container, and replace it with the sample to be measured, place it back in the measurement chamber, and make an unknown sample run.
- During the unknown sample run, the digital control system decreases the power in the calorimeter until the bridge voltage is the same as that measured in the zero baseline run.
- The power input to the calorimeter during this unknown sample run ( $W_{usr}$ ) is measured.
- The power of the sample being measured ( $W_s$ ) is calculated by subtracting the wattage value measured during the zero baseline run from the wattage measured during the unknown sample run to find the wattage of the sample. In equation form:

$$W_s = W_{zbl} - W_{usr}$$

The calorimeter factor for tritium used at most DOE sites for the purposes of reporting accountable quantities of tritium to DOE is 0.3240 +/- 0.0009 Watts/g of tritium.

CHF calorimetry can be used to measure tritium in solid form. CHF is the most accurate method available for the measurement of tritium quantities if the chamber size and wattage level of the item to be measured are well matched to the specifications of the CHF system being used. CHF systems, however

- Do not provide any information about the different gases present in a container (e.g., HT, DT, H<sub>2</sub>, D<sub>2</sub>, <sup>3</sup>He),
- Only measure the quantity of tritium in the container,
- Are not currently available for items larger than 11 inches in diameter and 16 inches long, and
- Take several hours to complete a single measurement.

## APPENDIX D: CONTAMINATION AND SURFACE ACTIVITY THRESHOLDS

### D.1 Appendix D to Part 835 - Surface Radioactivity Values

The data presented in Appendix D are to be used in identifying contamination and high contamination areas as defined in Sec. 835.2(a), identifying the need for surface contamination monitoring and control in accordance with Sec. 835.404, identifying the need for radioactive material controls in accordance with Sec. 835.1101.

Surface Radioactivity Values {1}  
[In dpm/100 cm<sup>2</sup>]

| Radionuclide                                                                                                                                   | Removable<br>{2}, {4} | Total (fixed +<br>removable) {2},<br>{3} |
|------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------|------------------------------------------|
| U-nat, U-235, U-238, and associated decay products                                                                                             | 1,000{7}              | 5,000                                    |
| Transuranics, Ra-226, Ra-228, Th-230, Th-228, Pa-231, Ac-227,<br>I-125, I-129                                                                  | 20                    | 500                                      |
| Th-nat, Th-232, Sr-90, Ra-223, Ra-224, U-232, I-126, I-131, I-133                                                                              | 200                   | 1,000                                    |
| Beta-gamma emitters (nuclides with decay modes other than alpha<br>emission or spontaneous fission) except Sr-90 and others noted above<br>{5} | 1,000                 | 5,000                                    |
| Tritium and tritiated compounds {6}                                                                                                            | 10,000                | See footnote 6                           |

{1} The values in this Appendix, with the exception noted in footnote 6, apply to radioactive contamination deposited on, but not incorporated into the interior of, the contaminated item. Where surface contamination by both alpha- and beta-gamma-emitting nuclides exists, the limits established for alpha- and beta-gamma-emitting nuclides apply independently.

{2} As used in this table, dpm (disintegrations per minute) means the rate of emission by radioactive material as determined by correcting the counts per minute observed by an appropriate detector for background, efficiency, and geometric factors associated with the instrumentation.

{3} The levels may be averaged over one square meter provided the maximum surface activity in any area of 100 cm<sup>2</sup> is less than three times the value specified. For purposes of averaging, any square meter of surface should be considered to be above the surface radioactivity value if (1) from measurements of a representative number of sections it is determined that the average contamination level exceeds the applicable value; or (2) it is determined that the sum of the activity of all isolated spots or particles in any 100 cm<sup>2</sup> area exceeds three times the applicable value.

{4} The amount of removable radioactive material per 100 cm<sup>2</sup> of surface area should be determined by swiping the area with dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency. (Note—The use of dry material may not be appropriate for tritium.) When removable contamination on objects of surface area less than 100 cm<sup>2</sup> is determined, the activity per unit area should be based on the actual area, and the entire surface should be wiped. It is not necessary to use swiping

techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination.

{5} This category of radionuclides includes mixed fission products, including the Sr-90 which is present in them. It does not apply to Sr-90 which has been separated from the other fission products or mixtures where the Sr-90 has been enriched.

{6} Tritium contamination may diffuse into the volume or matrix of materials. Evaluation of surface contamination shall consider the extent to which such contamination may migrate to the surface in order to ensure the surface contamination value provided in this appendix is not exceeded. Once this contamination migrates to the surface, it may be removable, not fixed; therefore, a "Total" value does not apply. In certain cases, a "Total" value of 10,000 dpm/100 cm<sup>2</sup> may be applicable either to metals of the types from which insoluble special tritium compounds are formed, that have been exposed to tritium, or to bulk materials to which insoluble special tritium compound particles are fixed to a surface.

{7} These limits apply only to the alpha emitters within the respective decay series.

D.2 Response to Questions and Clarification of Requirements and Processes: DOE Order 5400.5 Chg. 2, Section II.5 and Chapter IV Implementation (Requirements Relating to Residual Radioactive Materials), DOE Office of Assistant Secretary for Environment, Safety and Health, dated November 17, 1995

TABLE 1 SURFACE ACTIVITY GUIDELINES  
Allowable Total Residual Surface Activity (dpm/100 cm<sup>2</sup>)<sup>4</sup>

| Radionuclides <sup>5</sup>                                                                                                                                                     | Average <sup>6/7</sup> | Maximum <sup>9/8</sup> | Removable <sup>9</sup> |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------|------------------------|------------------------|
| Group 1 – Transuranics, I-125, I-129, Ac-227, Ra-226, Ra-228, Th-228, Th-230, Pa-231                                                                                           | 100                    | 300                    | 20                     |
| Group 2 – Th-natural, Sr-90, I-126, I-131, I-133, Ra-223, Ra-224, U-232, Th-232                                                                                                | 1,000                  | 3,000                  | 200                    |
| Group 3 – U-natural, U-235, U-238, and associated decay products, alpha emitters                                                                                               | 5,000                  | 15,000                 | 1,000                  |
| Group 4 – Beta-gamma emitters (radionuclides with decay modes other than alpha emission or spontaneous <sup>10</sup> fission) except Sr-90 and others noted above <sup>7</sup> | 5,000                  | 15,000                 | 1,000                  |
| Tritium (applicable to surface and subsurface) <sup>11</sup>                                                                                                                   | N/A                    | N/A                    | 10,000                 |

4 As used in this table, dpm (disintegrations per minute) means the rate of emission by radioactive material as determined by counts per minute measured by an appropriate detector for background, efficiency, and geometric factors associated with the instrumentation.

5 Where surface contamination by both alpha- and beta-gamma-emitting radionuclides exists, the limits established for alpha- and beta-gamma-emitting radionuclides should apply independently.

6 Measurements of average contamination should not be averaged over an area of more than 1 m<sup>2</sup>. For objects of smaller surface area, the average should be derived for each such object.

- 7 The average and maximum dose rates associated with surface contamination resulting from beta-gamma emitters should not exceed 0.2 mrad/ h and 1.0 mrad/ h, respectively, at 1 cm.
- 8 The maximum contamination level applies to an area of not more than 100 cm<sup>2</sup>.
- 9 The amount of removable material per 100 cm<sup>2</sup> of surface area should be determined by wiping an area of that size with dry filter or soft absorbent paper, applying moderate pressure, and measuring the amount of radioactive material on the wiping with an appropriate instrument of known efficiency. When removable contamination on objects of surface area less than 100 cm<sup>2</sup> is determined, the activity per unit area should be based on the actual area, and the entire surface should be wiped. It is not necessary to use wiping techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination.
- 10 This category of radionuclides includes mixed fission products, including the Sr-90 that is present in them. It does not apply to Sr-90 that has been separated from the other fission products or mixtures where the Sr-90 has been enriched.
- 11 Property recently exposed or decontaminated should have measurements (smears) at regular time intervals to ensure that there is not a build-up of contamination over time. Because tritium typically penetrates material it contacts, the surface guidelines in Group 4 are not applicable to tritium. The Department has reviewed the analysis conducted by the DOE Tritium Surface Contamination Limits Committee ("Recommended Tritium Surface Contamination Release Guides," February 1991), and has assessed potential doses associated with the release of property containing residual tritium. The Department recommends the use of the stated guideline as an interim value for removable tritium. Measurements demonstrating compliance of the removable fraction of tritium on surfaces with this guideline are acceptable to ensure that non-removable fractions and residual tritium in mass will not cause exposures that exceed DOE dose limits and constraints.