TECHNICAL CHALLENGES FOR DRY STORAGE OF USED FUEL FROM RESEARCH REACTORS IN CANADA

Jim Lian, Tracy Sanderson and Aaron Chiu
Atomic Energy of Canada Limited, Chalk River Laboratories
Chalk River, Ontario K0J 1J0, Canada

ABSTRACT

Research reactors have been operated in Canada to support nuclear R&D programs and medical isotope production. Used nuclear fuel discharged from these research reactors is currently being dry stored at the storage facilities at Chalk River site. The research reactor used fuels are significantly less corrosion-resistant than the Zircaloy-clad power-reactor fuels during storage and are subject to unique hazards associated with their chemical and physical characteristics. This paper discusses the technical and safety issues related to the dry storage of these fuels, and presents the key safety considerations required in the design and operation of new dry fuel storage facilities at CRL.

1. INTRODUCTION

The fleet of research reactors in Canada consists of eight operating reactors and six shut-down reactors located at various nuclear laboratories, universities and research centers across the country. The oldest research reactor (NRX) went to critical in 1947. Over the years, a considerable amount of used fuel discharged from these research reactors has being consolidated and managed at AECL's Chalk River Laboratories (CRL) site.

The used fuel inventory consists of dozens of types and configurations, and includes intact and non-intact fuels. The fuels are a mixture of both high and low enrichments, and are typically aluminium-clad with various fuel core materials, including uranium metal, uranium dioxide, uranium-aluminium alloy, and uranium silicide-aluminium dispersion fuels.

These used fuels are initially stored in water-filled fuel storage bays for a period of several years before being placed in storage containers and transferred to the CRL Waste Management Area "B" (WMA "B"), where they are stored in below-grade, vertical cylindrical steel and concrete structures called "tile holes". Tile holes storage structures are engineered to shield the radioactivity of the used fuel, dissipate heat and to contain any contamination, and thereby prevent the spread of radioactivity into the environment.

Compared with the Zircaloy-cladded power-reactor fuels, the aluminum-cladded Research Reactor Used Nuclear Fuels (RRSNF) are significantly less robust, less corrosion-resistant and can be subject to additional hazards (e.g., combustion and explosion) associated with their chemical and physical characteristics. Severe corrosion and degradation were reported in the US and other countries with research reactor used fuels stored under less-than-appropriate conditions. At CRL, some of the legacy fuels loaded in the older tile holes during the 1960's and 1970's, are also known to be degrading due to ingress of moisture.

In addition to using improved design of tile holes that are resistant to moisture ingress, AECL is in the process of designing and implementing new above-ground dry storage facilities to store the legacy fuels that will be retrieved from the older tile holes, and to accommodate future used fuel as well as other types of radioactive wastes generated at CRL. Figure 1 shows one such new facility, the Fuel Packaging and Storage (FPS) facility at CRL.

It is imperative that valid and realistic safety criteria be incorporated in the design and operation of these new dry storage facilities to ensure the safe and retrievable storage of the fuel over the designed storage life.

2. KEY SAFETY ISSUES WITH DRY STORAGE OF USED AL-CLAD FUEL

The fundamental requirements of a dry storage facility for used fuels are to maintain:

- Subcriticality,
- Containment of the radioactive materials,
- Adequate radiation shielding,
- Effective and reliable decay heat removal to keep the temperature of fuel and storage, structure below their limits, and
- Retrievability of the used fuel at the end of storage life for final disposal.

These design requirements are applicable to both wet and dry stores, and are common to both power-reactor used fuel and research-reactor used fuel. Detailed discussions of these design requirements (subcriticality, heat removal, shielding, containment and retrievability) are given in Reference 1. This paper focuses on issues specific to dry storage of aluminum-cladded RRSNF, which may be subject to aggressive corrosion and subsequent safety hazards under improper storage conditions.

In most cases when dry storage is considered, interim storage containers are not foreseen to be used for final disposal. Therefore at the end of storage life, the RRSNF would need to be retrieved safely from interim dry storage for re-conditioning and re-packaging prior to its emplacement in the final disposal facility.

Retrievability of the stored used fuel is related directly to limiting degradation of the storage system. Excessive degradation would distort the configuration and reduce the mechanical strength of the stored fuel assemblies to a degree that may compromise retrievability.

Degradation of the RRSNF cladding and fuel by corrosion occurs from exposure to air and/or sources of humidity. The presence of excess humidity or water in the fuel container would result in excessive corrosion of the fuel assemblies during storage leading to:

- Poor retrievability due to loss of fuel integrity and excessive distortion of fuel assemblies,
- Impaired containment as a result of cladding breach,
- Risk of fire/explosion due to generation of hydrogen gas from fuel corrosion and water radiolysis, and generation of pyrophoric materials, and
- Increased risk of loss of subcriticality due to the presence of water and loss of the original configuration of fuel assembly.

For this reason, the vulnerability to degradation and associated difficulties in retrieval are considered as the two major issues specific to dry storage of RRSNF.

2.1 Degradation of the Fuel Aluminum Cladding

The potential degradation mechanisms of used Al-clad fuel during dry storage were evaluated by US DOE Savannah River National Laboratory [2]. The range of environmental conditions considered are temperatures less than 600°C; cover gases of air, non-reactive, or inert gas; and relative humidity levels up to 100%. The potential degradation mechanisms identified and evaluated include:

- Corrosion of the cladding and fuel from exposure to air and/or sources of humidity;
- Dehydration of the hydrated oxide films with subsequent corrosion of the cladding,
- Hydrogen blistering of the aluminum cladding,
- Radiation embrittlement of the cladding,
- Inter-diffusion of the fuel and fission products with the cladding, and
- Creep of the cladding due to self-weight loading and loading from swelled fuel.

It was concluded in [2] that these mechanisms, except for corrosion, are only significant at temperatures well above 200°C.

For the dry storage facilities of used Al-clad fuel, the maximum fuel temperature should be 200°C to prevent occurrence of the above degradation mechanism other than corrosion. Therefore, in the presence of water vapour and/or air, corrosion of the aluminum cladding and the fuel matrix materials is the most limiting mechanism that could cause significant degradation of the fuel during dry storage.

2.2 Corrosion of Exposed Fuel Core Materials

When a breach of the aluminium cladding occurs, the fuel core material is exposed to the corrosive environment. The behaviour of the exposed fuel core material depends on the types of the fuel material.

2.2.1 Corrosion of Exposed Uranium Metal

The uranium metal is very reactive chemically. It reacts actively with water (liquid or vapour) and every major atmospheric component [3]. The reaction with water is considered the primary cause of corrosion of the exposed uranium metal under conditions relevant to dry storage. The products of uranium-water reaction are uranium oxide and hydrogen gas. Under oxygen-depletion and hydrogen-rich conditions, such as those that can occur in a sealed storage container, the molecular hydrogen generated is dissociated by uranium and forms uranium hydride (UH_3). The uranium hydride, a loose fine black particulate, is pyrophoric at room temperature when exposed to air.

Due to the high reactivity of the uranium metal, fuel with exposed uranium metal would need to be thoroughly dried and stored in dry inert cover gas.

The release of fission products follows the progress of uranium oxidation; there is no leaching of fission products from un-reacted uranium metal in water [3]. In other words, the fission product inventories within the un-corroded uranium metal would remain un-released.

2.2.2 Corrosion of U-Al and Uranium Silicide-Aluminium Dispersion Fuels

The uranium-aluminium fuel and the uranium silicide-aluminium dispersion fuel are very much less reactive than uranium metal [3]. The resistance of the uranium-aluminium alloy to general corrosion is reported to be similar to that of the aluminium cladding [2].

When aluminium cladding of the uranium-aluminium fuel or the uranium silicide-aluminium dispersion fuel is penetrated, the immediate fuel core area at the point of breach is exposed and only the fission gas stored in the corroded fuel matrix would be available for release. The majority of fission products will be retained insider the fuel core.

Formation of pyrophoric uranium hydride from corrosion of the uranium-aluminium fuel is not considered as a concern in any of the references reviewed. The U_3Si differs from most uranium alloys in that it does not form a hydride [4].

2.3 Potential Hazards of Severe Damage to the Fuel Storage Container

Internal events, such as fire or explosion or over-pressurization of a sealed container can result in severe damage to the fuel storage system, including fuel storage containers. For a dry storage facility containing used Al-clad research reactor fuel, the potential hazards that can lead to such damages are: (1) fire and explosion due to hydrogen build-up or sealed container over-pressurization due to gas generation, and (2) fire and explosion due to formation of uranium hydride.

2.3.1 Hydrogen Build-Up

At temperatures above approximately 80°C, hydrogen build-up occurs in a closed system containing aluminum and water according to [5]:

$$2\,Al + 4\,H_2O = Al_2O_3 \bullet H_2O + 3\,H_2$$

The generation of hydrogen in the reaction to produce Boehmite ($Al_2O_3 \bullet H_2O$) bounds that for the reaction to produce Gibbsite ($Al_2O_3 \bullet 3H_2O$) which occurs at temperatures below approximately 80°C.

One impact of hydrogen build-up is the potential for an explosion hazard. The lower concentration limits are:

- 4% by volume in air at room temperature for flammability of hydrogen
- 9% by volume in air for a sustained burn of hydrogen, and
- 18% by volume for hydrogen/air mixture being explosive.

If an inert gas is used as the cover gas for the fuel storage container containing dried Al-clad fuel, the potential to produce a flammable mixture would be reduced or eliminated.

In addition to being a fire/explosion hazard, accumulation of excessive hydrogen gas may also over-pressurize the sealed storage container, and cause hydrogen embrittlement of the container materials at locations of high stress and surface discontinuities over long storage times [6].

2.3.2 Generation of Pyrophoric Uranium Hydride

Generation of pyrophoric uranium hydride is a concern only for dry storage of uranium metal fuel. When the aluminium cladding is breached, the metallic uranium may be exposed to water or water vapor left over within the sealed container, leading to the following reaction [7]:

$$U + 2\ H_2O \rightarrow 2\ UO_2 + H_2$$

The hydrogen will react with the uranium to form uranium hydride according to the reaction:

$$2\ U + 3\ H_2 \rightarrow 2\ UH_3$$

The rate of this reaction is slow, and only occurs under oxygen-depletion and hydrogen-rich conditions. The production of hydride will stop once the oxygen content of the humid atmosphere in contact with the fuel is raised above 100 mg/kg.

If oxygen is present in the container, it reacts quickly with the uranium hydride formed to produce more water and uranium oxide according to the reaction:

$$2\ UH_3 + 3.5\ O_2 \rightarrow 2\ UO_2 + 3\ H_2O$$

In a closed system initially filled with humid air, the above reactions will continue until all the oxygen, water and hydrogen are used up. The third reaction will be the final reaction, due to the oxygen-depletion limit, so that the hydride still remains at the end state. The system pressure will be below atmosphere pressure because the oxygen is removed from the gas phase.

This presents a potential explosion hazard for fuel retrieval at the end of interim dry storage. The loose uranium hydride powder deposited on the surface of the exposed uranium metal fuel would be disturbed and may suspend in the air when the fuel storage container is handled during retrieval. When the fuel container closure is opened, fresh air would rush into the container due to the negative container pressure. A violent reaction of the oxygen in the air with the suspended uranium hydride may cause an explosion [7].

3. SPECIAL REQUIREMENTS FOR DRY STORAGE OF RRSNF

The above discussion shows that the aluminum-clad RRSNF is vulnerable to degradation during long term dry storage. Under improper storage conditions, excessive fuel degradation may occur and compromise criticality safety, containment, shielding and retrievability of RRSNF during long term dry storage.

In addition to the generic requirements for dry storage of used nuclear fuel (such as criticality safety, radiation shielding, decay heat removal, etc.) the following specific requirements for the new dry storage facilities for RRSNF at CRL are proposed:

1. The maximum fuel temperature during handling (e.g., drying) and storage will not exceed 200°C.

   Degradation of the fuel is a temperature-dependent process. By limiting the fuel temperature below 200°C, degradations by hydrogen blistering, inter-diffusion of the fuel and fission products with the cladding, radiation embrittlement and creeping become insignificant. The only remaining degradation mechanism of significance is corrosion.

2. The used fuel must be adequately dried prior to storage to further minimize fuel corrosion during long term storage.
3. After drying, the used fuel must be maintained in a dry and non-reactive or inert cover gas environment, which minimizes fuel corrosion and reduces or eliminates the potential hazards of fire/explosion due to build-up of hydrogen gas from corrosion and due to formation of pyrophoric corrosion products.

4. KEY DESIGN CONSIDERATIONS FOR DRY STORAGE OF RRSNF AT CRL

For the new dry storage facilities at CRL, the used fuel assemblies will be loaded into a storage container and then dried to remove the residual water. At the completion of the drying process, the loaded storage container will be sealed, vacuumed and then backfilled with inert cover gas. Figure 2 shows a general arrangement of a storage location.

Among the three special requirements discussed in Section 3, the fuel temperature limit of 200°C can be assured by the design of the passive heat removal system of the storage facilities under normal, abnormal and credible accident conditions. Evacuating and back filling of the loaded storage container with inert gas can also be accomplished with no anticipated technical difficulties.

However, it is the adequacy of the fuel drying process that continues to be a key technical issue with the dry storage of RRSNF. The issue is of two folds: (1) how dry is considered adequately dry? (2) if the dryness criterion is specified, how to demonstrate that the criterion is met when the drying process is completed?

4.1 Adequacy of Fuel Drying

The purpose of the drying process is to remove residual water from the fuel storage container before backfilling the sealed container with inert gas for long term dry storage. Vacuum drying combined with (hot or cool) gas purging is generally considered to be the best option to remove residual water from used fuel retrieved from pool storage. A drying acceptance criterion must be established so that the operator can determine when the fuel is sufficiently dried and to terminate the drying process.

For a standard vacuum drying processes, adequate water removal is normally evaluated using vacuum pressure rebound measurements. The pressure rebound measurements consist of showing that an evacuated container loaded with used fuel will retain vacuum for a specified period without a pressure rise greater than a specified limit, indicating that any free liquid water on the fuel has been vaporized and evacuated.

The residual water that may exist in a used fuel container can be in the forms of free water, physisorbed water and chemisorbed water. A proper vacuum drying process for RRSNF can remove the free water and most of the physisorbed water, but would be ineffective in removing the chemisorbed water. For RRSNF with aluminum cladding, the chemisorbed water exists in the surface hydroxides, including Boehmite (Al2O3•H2O). Boehmite remains stable to temperature >340°C, much higher than the recommended temperature limit of 200°C allowed for the aluminum clad RRSNF during fuel handling and storage.

The amount of chemisorbed water in the form of Boehmite could be significant. For example, an average of approximately 1.7 L of Boehmite water content is expected to exist in a storage canister containing 30 used fuel assemblies of US Advanced Test Reactor (ATR) [6]. Exposure of Boehmite to gamma irradiation can produce radiolytic hydrogen gas [8]. In other words, it would still be necessary to demonstrate that the accumulation of hydrogen gas due to radiolysis of Boehmite would not be an issue for a closed storage container during long term storage, even after a rigorous drying process.

For the legacy fuel at CRL, fuel drying presents a particular challenge because the existing packaging of these fuel assemblies (e.g., re-cans or closed cans) allows very restricted pathways for the humidity flow during the vacuum drying process. This would make the drying process less effective, and more importantly, it provides no assurance of fuel dryness even if the vacuum pressure rebound measurements appeared satisfactory due to the very restricted pathways for the humidity flow. The amount of water existing in the packages retrieved from the old tile holes varies from package to package, and is often unknown. Following the drying process, the amount of residual water within the storage container remains unknown, providing no basis for demonstrating that the fuel has been adequately dried.

A sealed storage system is particularly vulnerable to inadequate drying, which may result in continuous corrosion of the stored fuel and increased risks of fire/explosion or over-pressurization from accumulation of hydrogen gas and other pyrophoric materials.

4.2 CRL's Approach

In recognizing that the RRSNF at CRL, especially the legacy fuel assemblies retrieved from the older tile holes, may not always be adequately dried prior to dry storage, a vented storage system is selected to accommodate the potential issues resulted from inadequate drying process.

A vented system stores used fuel assemblies in non-sealed containers or holders that are accessible to the environment of the facility. The dry storage facilities are monolithic concrete vaults housing a number of storage wells, as shown in Figure1. In each of the storage well, the used fuel is contained inside a stainless steel storage container (Figure 2). The top of the storage container is fitted with metal filters to allow venting of container while prevent radioactive particulate from escaping. The top of the storage well is closed by a shield plug which is equipped with two vent ports to facilitate vacuum drying, purging and backfilling with cover gas, as well as gas sampling.

Vented systems have the advantage that residual water in and around the used fuel can evaporate or radiolyze over long time and escape from the system. However, exposure of the container to ambient air also allows ingress of moisture and impurities in the air by aspiration of humid air from the external atmosphere. The entered moisture may condensate and accumulate in the container causing corrosion of the used fuel and container. To prevent ingress of moisture and impurities the vent ports of the storage wells are normally kept closed once the vacuum drying is complete, and are opened only when conducting periodic inspection and sampling.

The main features of using the proposed vented storage system are:

- The used fuel will be vacuum dried to remove as much residual water as practical and then back filled with dry inert gas.
- The designer accepts the fact that: (1) there will still be certain amount of residual water left after drying, and (2) the residual water/humidity would lead to fuel corrosion and hydrogen gas generation within the storage well, which is normally closed by valves but can be vented as necessary.
- The corrosion of the aluminum cladding and the build-up of hydrogen gas progress at slow rates, allowing sufficient time for detection and remedial actions.
- The pressure within the storage well is monitored and sampled periodically. Higher pressure and elevated hydrogen gas concentration in the storage well indicate excessive residual water.
- In-situ gas purging, re-drying and re-filling of inert gas will be provided for storage locations in which excessive residual water is detected.

- Multiple barriers of containment[1] of radioactive materials in the used fuel are provided:
  - By the cladding of the RRSNF. In the event of cladding failure, very little fission gas will be released as the majority of fission products will be retained insider the fuel matrix (see section 2.2).
  - By the metal filters that confines any radioactive particulates released from the fuel.
  - By the normally closed storage well assembly. Any released fission gases will be contained in the storage tube headspace, which can be vented in a controlled manner.
- The vented design facilitates the monitoring, purging, re-drying and inert gas re-filling processes to minimize exposure of the staff.

5. SUMMARY

The majority of used fuels discharged from research reactors in Canada are stored at AECL's Chalk River site. The Al-clad research reactor used fuels are prone to degradation during long term storage under improper storage conditions. Excessive degradation of the fuel and fuel packages may compromise the criticality safety, containment and retrievability of the stored fuel and pose risks of fire/explosion to the facility.

The rate and extent of fuel degradation during dry storage are very sensitive to the storage environment conditions. To minimize fuel degradation and the associated risk of an energetic event, a sealed storage container containing RRSNF must be adequately dried to remove the residual water/humidity and then backfilled with a non-reactive dry gas, the fuel temperature must also be maintained below 200°C throughout the dry storage period.

For some of the legacy fuels to be stored in the new dry storage facilities at CRL, the characteristics of these fuels in their existing packaging make it difficult to both achieve and demonstrate adequate fuel drying. To accommodate these difficulties, a controlled-vented storage system, instead of the sealed storage system, is selected.

Prior to storage, the used fuel will be dried rigorously to remove as much residual water as practical, and then stored in an inert gas environment. Subsequent release of residual water in the normally-closed vented system can be detected by a monitoring and inspection system of the storage gas via the venting ports. If excessive residual water is found in any of the storage locations, the storage space of these locations can be purged, re-dried and re-filled in-situ.

The vented storage system is feasible for dry storage of the research reactor used fuels because:

- Both the fuel cladding and the fuel matrix provide containment of the fission products. Hence the risk and consequences of radiological release is significantly lower than that of power-reactor used fuel. There will be no burst release of fission gases upon cladding failure, as most of the fission products are still retained inside the fuel matrix.
- No release of radioactive particulates, which are contained by metal filters.

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[1] A barrier is defined as a natural or engineered feature which delays or prevents material migration to or from storage components [1].

- Radioactive gases are confined within the system which is normally closed by valves, and can be vented only in a well controlled manner.
- Fuel cladding corrosion is a slow process at the storage temperature allowing sufficient time for detecting and mitigating actions.
- As the new dry storage facilities are above-ground and shielded structures, and the vented storage system is normally closed, ingress of water/humidity into the storage wells is essentially eliminated. Any residual water, if it exists in the storage well after drying, would be depleted in the first few years of storage, ensuring a dry environment over the longer term.

6. REFERENCES

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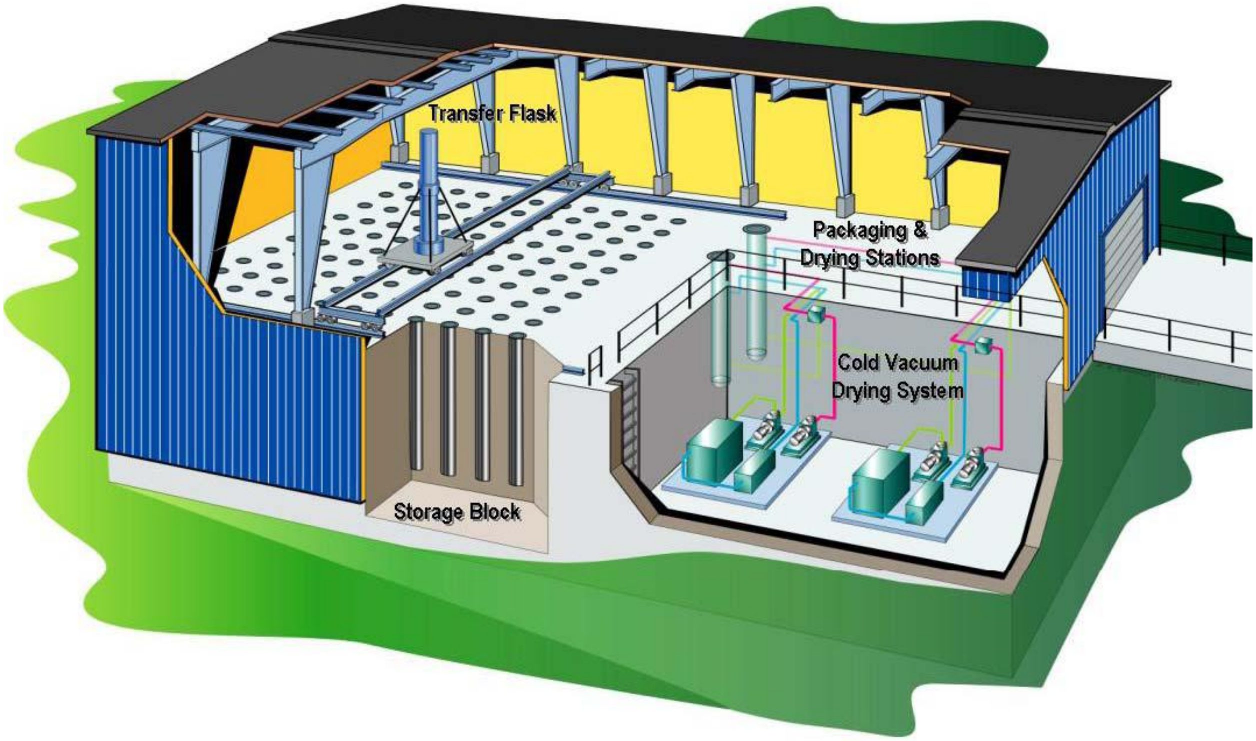

Figure 1. Fuel Packaging and Storage Facility

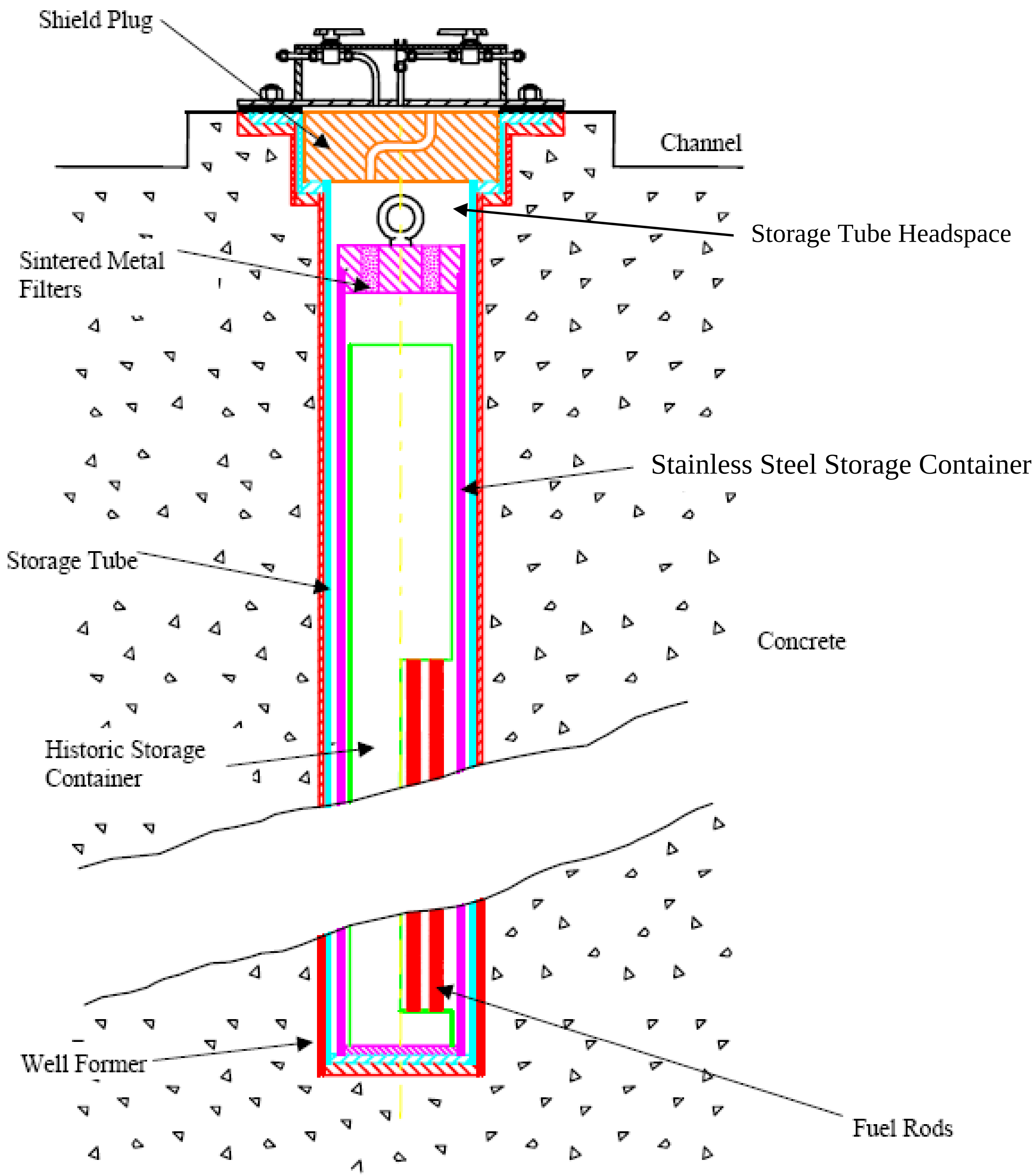

Figure 2. Arrangement within a Loaded Fuel Storage Location