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Melt-Dilute Treatment of Russian Research and Test Reactor Fuel Feasibility Study

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1.0 EXECUTIVE SUMMARY

This report provides a detailed overview of the melt-dilute treatment technology for aluminum-clad, aluminum-based fuels. The melt-dilute treatment technology has been developed at the Savannah River Site to reduce the volume and uranium enrichment of these fuels for the purpose of producing a waste form for ultimate disposal. This report provides an evaluation of the feasibility of adapting the melt-dilute treatment for the complete inventory of spent nuclear fuel from research and test reactors owned by Russia. The inventory of fuels from these reactors include aluminum-clad, aluminum-based fuels, plus other cladding and fuel meat materials that could potentially be treated to achieve a monolithic ingot suitable for long-term storage or disposal.

The basic steps to produce and package a melt-dilute waste form are described. In the treatment of aluminum-clad, aluminum-based fuels that contain high-enriched uranium, a melt process is prescribed to achieve isotopic dilution via additions of depleted uranium to a melt targeted at the eutectic composition of the uranium-aluminum alloy. The eutectic composition minimizes the required temperature of the melt and thus species that would be produced in the off-gas stream. Melt treatment prescriptions for additions of catalyst species to reduce oxide and intermetallic compounds of uranium to enable dilution are described. Melt treatment prescriptions for additions of neutron absorbers species, that may be required to ensure criticality safety in disposal systems, are also described.

The Russian spent fuels contain predominantly moderate- or low-enriched uranium. These may not need isotopic dilution in the treatment process used to produce a consolidated form, and therefore the treatment process may be simplified. Additional development of melt treatments would be needed for several cladding and fuel meat combinations in the Russian fuel inventory. Basic strategies for this development of the melt treatment for each of the types of Russian fuel are described.

Design concepts for a melter, an off-gas system, secondary waste disposal, and fuel/waste form handling and packaging are described. Additional tests are described that were done using a closed system to eliminate the need for an off-gas system. Analyses to meet potential disposal system acceptance criteria for waste forms such as the avoidance of criticality over geologic time periods, and the dissolution/reconfiguration of the waste form under environments projected for the disposal system are also addressed. This is performed using the acceptance criteria for a melt-dilute waste form for the U.S. Mined Geologic Repository as an example.

The melt-dilute treatment facility is amenable to a transportable, modular design, and a design concept is outlined in this report.

2.0 BACKGROUND

The U.S. Department of Energy is collaborating with Russia to reduce proliferation challenges posed by weapons-usable nuclear material. Activities between the two countries reflect their concern over the risk of theft and diversion of these materials. Joint research and development efforts between the countries are directed at enhancing proliferation resistance by focusing on the development of nuclear disposal options that maximize technological proliferation barriers.

One source of weapons-usable material is spent research reactor fuel assemblies that are stored at various reactor sites around the world. The U.S. has a program to bring back to the Savannah River Site (SRS) spent research reactor fuel of U.S. origin that have been sent as reactor fuel to foreign countries. The SRS plan is to consolidate the fuel and put the material in a form that is safe to store and is proliferation resistant. A treatment option that is an alternative to reprocessing is the Melt-Dilute (MD) technology. This technology simply melts the spent nuclear fuel (SNF) and adds depleted uranium to dilute the isotopic uranium content to less than 20% ^{235}U . A similar take back program is proposed for spent Russian research reactor fuel that has been sent to satellite countries. The MD technology, especially a developed mobile MD treatment unit, may be applicable for treatment of this fuel.

Initially, Russian fuel assemblies were fabricated using highly enriched uranium (HEU). Recently, they converted to lower enriched uranium for manufacturing research reactor fuel elements, but many of the Russian HEU spent fuel assemblies are stored in water storage pools at various reactor sites. The IAEA along with Russian scientist are expecting the return of spent fuel assemblies to Mayak in the former Soviet Union for storage and final disposition. A team of U.S. and Russian experts recently accompanied the IAEA in an inspection of the spent fuel at several reactor storage sites. They reported mixed fuel conditions at these sites [1].

The melt-dilute treatment process for aluminum-clad, aluminum-based spent fuel was developed as an alternative process at the Savannah River Site for ultimate disposition of foreign and domestic research reactor fuels being consolidated at the site. The process consists of the melting of the spent nuclear fuel assembly and addition of depleted uranium to dilute the isotopic uranium content to less than 20%, the proliferation limit. Aluminum is added, if needed, to keep the melt composition near 13.8 wt% uranium, the eutectic composition. The eutectic composition is the lowest liquidus temperature in the uranium-aluminum alloy system. Once solidified, the ingot would be loaded into a special-design canister destined for disposal in a geologic repository. Because the isotopic content is low, criticality concerns are reduced and proliferation resistance is increased.

There are three primary categories in the SRS inventory of aluminum-clad, aluminum-based fuel: 1) Al-clad UAl_x fuel, 2) Al-clad uranium silicide powder metallurgy fuel, and 3) Al-clad U_3O_8 powder metallurgy fuel. These fuel categories each have a defined melt-dilute treatment process. Table 1 in the Appendix provides a listing of the Russian origin fuels. The three aluminum-clad, aluminum-based fuel categories cover the majority of the Russian fuels.

The following sections of this report provide a description of the steps in the melt-dilute treatment of aluminum-clad, aluminum-based fuel. A brief discussion of the development needs for treating the other types of Russian fuels is also made.

3.0 SPENT FUEL INVENTORIES

Russia's research and tests reactor spent nuclear fuel inventory contains aluminum clad fuel assemblies with a mixture of fuel core materials, including uranium-aluminum alloy, U_3O_8 -aluminum or UO_2 -aluminum, and some UO_2 mixed with magnesium. There are small quantities of uranium metal fuel and uranium-silicide fuel clad with aluminum and uranium-chromium-nickel alloy rods clad with stainless steel. Also, there are some uranium-zirconium fuels clad with zirconium. A list of the elements by type is listed in Table I of the Appendix along with uranium loading, quantity and enrichment. The majority of the spent fuel assemblies in the table were fabricated using either medium enriched uranium (MEU) or low enriched uranium (LEU) fuel. The average burnup is about 30-50%.

There are about 19,000 research assemblies identified in the list that contain approximately 500 metric tons of enriched uranium. The Russian fuel assemblies are similar in size to the Material Test Reactor (MTR) type research reactor fuel developed in the US. The fuel elements are either square, approximately 71x71 mm, or hexagonal in shape and are about 60 mm high.

4.0 MELT-DILUTE TREATMENT PROCESS AND HANDLING STEPS

The MD treatment process is a batch process that involves melting several spent fuel assemblies and adding depleted uranium to dilute the melt isotopic content to less than 20%, the proliferation limit. Because the isotopic content is low, criticality concerns are also reduced. The process for aluminum-based spent nuclear fuel is discussed below. Aluminum is added, as necessary, to keep the melt composition near 13.8 wt% uranium, the eutectic composition to minimize the process temperature. The eutectic composition provides the lowest liquidus temperature in the uranium-aluminum alloy system. Low operating temperatures reduces fission product volatility as well as enhances furnace and crucible life. Once solidified, the ingot is a wasteform that is put into a canister for long-term dry storage or ultimate disposal in a geologic repository. The MD treatment process was originally developed for the aluminum-based fuel systems to be received at SRS, but it is feasible to extend to other aluminum-based fuel systems as described below.

4.1 Melt-Dilute Process Steps

The general process flowsheet for the Melt-Dilute treatment technology are shown in Figure 4-1. These treatment steps depict the flow of spent nuclear fuels through full-scale production facility. A sub-set of these treatment steps—namely, the Melt-Dilute Treatment and Post-Treatment Characterization-- can also be performed using a modular, transportable melt-dilute system.

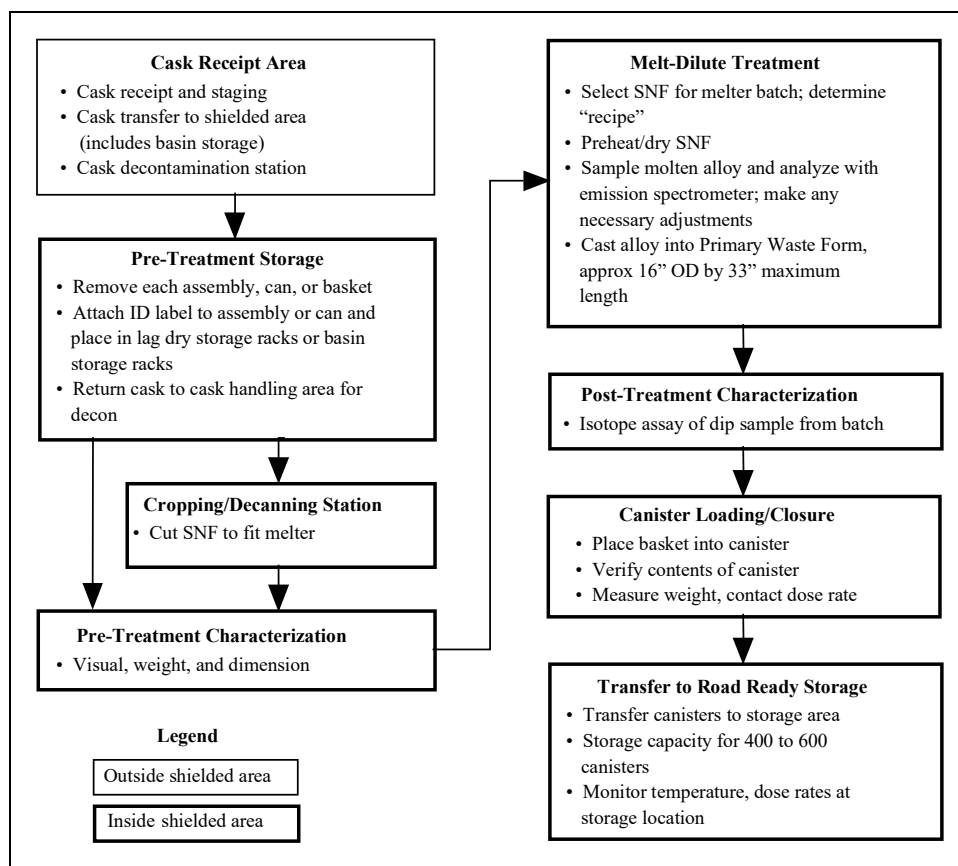


Figure 4-1 Process Steps for Melt-Dilute Treatment Using a Fixed or Mobile, Modular Facility

4.2 Mobile Melter Facility

A Mobile Melt-Dilute facility (MMD) is proposed for treatment of fuels at several storage locations in Russia; thereby, avoiding the costs of building separate treatment facilities at each site and avoiding shipment of enriched fuel assemblies over the road. The Mobile Melter facility concept is based on SRTC tests, and modular pilot-scale facilities constructed at SRS for treatment of U.S. spent fuel. Laboratory tests at SRTC have shown the feasibility of operating both a closed and a filtered off-gas system.

The laboratory tests for a closed system approach was done in 1997 by first evacuating a cylindrical vessel containing simulated fuel with various fission products. The test apparatus, shown in Figure 4-2, consisted of a tube that was first evacuated and sealed. Any pressure increase during melting due to residual air or volatile elements was measured using the pressure gage. The tube was heated using a resistance furnace. After heating at 850 °C for 1 hour, the test chamber was removed, cooled and opened. A white powder was observed near the top surface of the chamber where cesium and other simulated volatile fission products had condensed. The white powder, most likely cesium metal, oxidized upon exposure to air. However, if the container were used for spent fuel storage, it would not be opened, so the ingot and all volatile fission products would remain inside, thus eliminating the need for the filtered off-gas system. Upon cooling the gas pressure inside returned to approximately its original level.

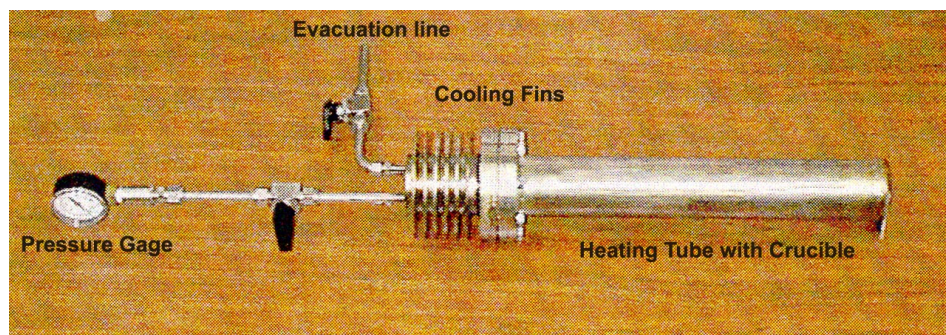


Figure 4-2 SRS Test Apparatus for Closed Melt-Dilute System Tests

One concept for the MMD, based on the above studies, would be to build a facility that would utilize the closed system approach. A schematic of such a system is shown in Figure 4-3 for induction heating. Because induction coils require water cooling, it may be advantageous, for simplicity and safety, to use resistance heating.

The furnace would be enclosed with an outer container or dome, similar to a reactor dome, to contain any volatile gases in the unlikely event the closed container leaked during melting. The dome may be designed to provide sufficient shielding so additional shielding may not be needed around the furnace or area.

Ideally, only one element would be melted at a time, but the system could be designed to melt 4-6 elements per batch. With one element the recipe for dilution and alloy composition control would be easier. It is expected that the furnace and controls would be located in separate international shipping containers that could be easily loaded onto trailers and unloaded at the work site for assembly.

Loading and unloading of the furnace with spent fuel would be done remotely. Once the fuel assembly is brought to the MMD facility in a cask, it would be unloaded using a forklift or crane and placed onto a remote system to move it to the furnace. It is expected that the cask would provide radiological shielding while furnace loading, unloading and transporting the spent fuel.

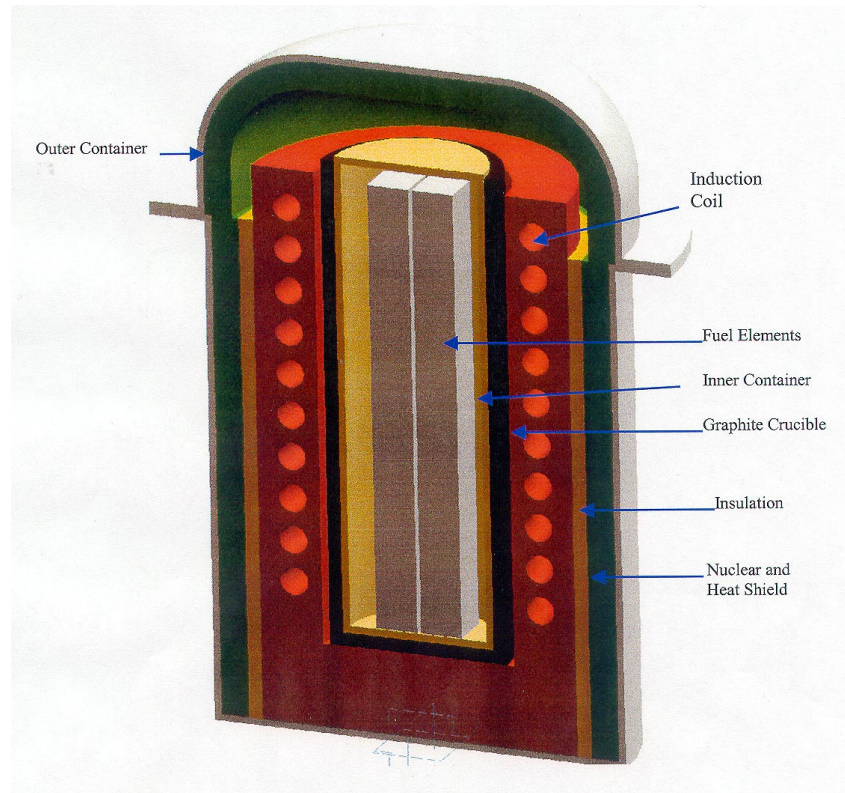


Figure 4-3 Induction Furnace for the Mobile Melt-Dilute Concept

The Mobile Melter can be transported in two over-the-road trailer assemblies. This allows free movement between locations where spent fuel is stored, eliminating the need to transport fuel and minimizing fuel handling. Only the melter and control system will move along roadways, it is not necessary to move shielding or structures. If additional shielding for process activities is needed, it can be easily erected at the individual sites. In addition, the control and monitoring equipment are housed separately keeping the more costly equipment in a clean environment at all times. Should contamination of the melter become a problem, the furnace can be replaced at minimal cost.

The sketch below, Figure 4.4, shows how the equipment could be installed in shipping containers and positioned at the storage facility. Inexpensive shielding, if needed, can be erected quickly and inexpensively with corrugated steel panels like those used to protect aircraft from strafing or ground attack. These panels can be erected to form cavities of predetermined thickness and filled with dirt. Control and power cables which must pass from the control unit to the melter can be installed through these cavities before they are filled, preventing "shine" along their paths. This

shortens the setup time required to make the system operational, leaving only three things to do: position the trailers, connect the cables, and test the system.

The melter was originally conceived as a closed system using a condenser to trap volatilized material, but an offgas system was added to avoid a pressurized system containing radionuclides and to reduce the waste volume. In addition to the closed system, a system similar to the one developed at SRS could be designed for mobility. Such a system would reduce waste volume up to 70%.

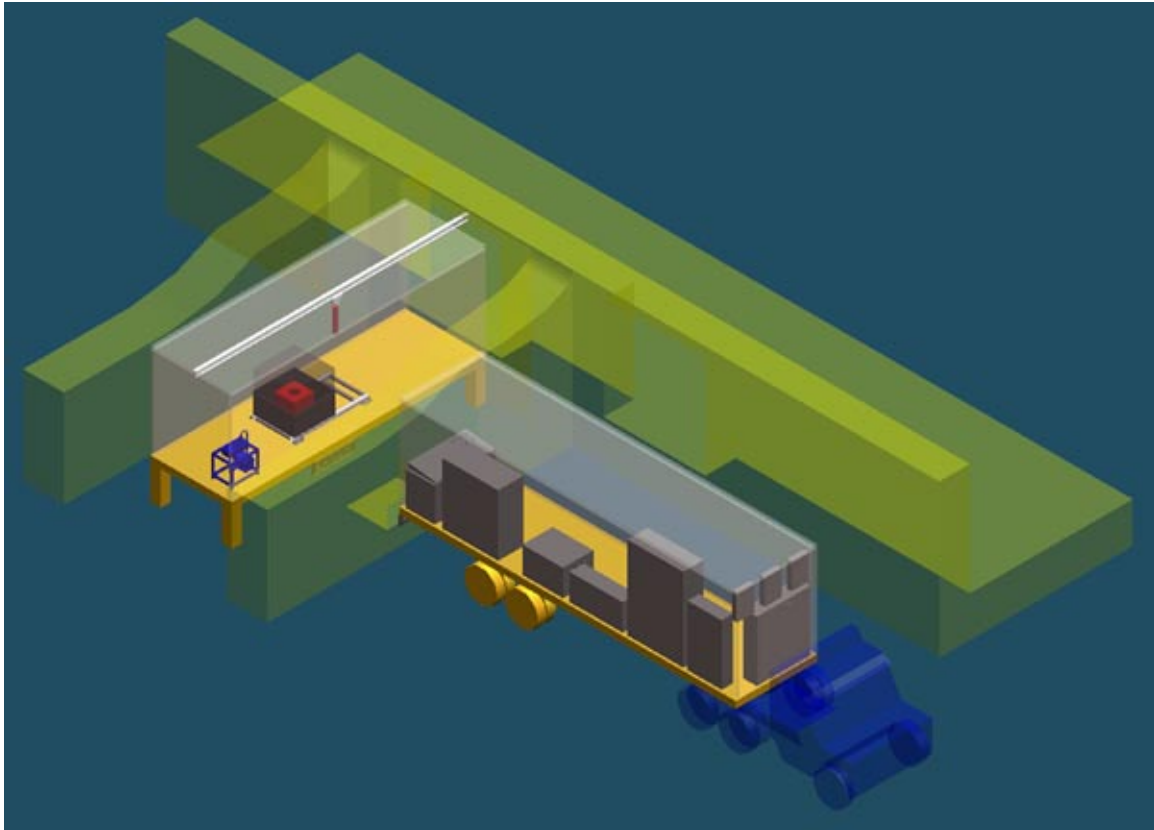


Figure 4-4 Modular Transportable Melt-Dilute System

5.0 MELT-DILUTE TREATMENT PROCESS

This section provides details of the overall treatment process. Included are descriptions of the waste form composition including neutron absorber additions, melter system design, off-gas system design, and secondary waste. Regulations for the U.S. facilities are cited.

5.1 Composition of the Melt-Dilute SNF Form

Significant research and development effort has been directed at the demonstration of the melt-dilute treatment of three fuel types. These include the binary uranium aluminum alloy fuels, the uranium silicide alloy fuels, and the uranium oxide alloy fuels. The first step in the development of the melt-dilute treatment process was to determine an appropriate alloy composition(s) since many processing variables such as temperature and time are highly dependent on composition. This section discusses the alloy compositions considered for the three fuel types.

Initially, three alloy compositions were investigated for the melt-dilute waste form. They are the eutectic composition, 30 wt% uranium-aluminum, and the intermetallic composition of UAl_4 . Primary focus was given to the eutectic composition because it has the lowest liquidus temperature in the U-Al alloy system. The 30 wt% uranium alloy has a liquidus temperature of less than 1000°C, and UAl_4 has the lowest liquidus temperature of the intermetallic compounds.

The eutectic composition contains about 87 wt% aluminum and 13 wt% uranium. Aluminum must be added to the melt for diluted alloy compositions above the eutectic. Aluminum adds about 370 cc/Kg to the waste volume, whereas uranium adds only about 50 cc/Kg. However, solidification of the eutectic composition is unique in that it occurs at a constant temperature of 646°C. The solidification characteristic of the eutectic produces a uniform microstructure with little or no gravity segregation of the aluminide intermetallic phase. Because the composition range is narrow, the microstructure generally consists of a small amount of primary aluminum or UAl_4 .

The liquidus temperature of the 30 wt% uranium composition is about 1000°C, and there is a 350°C two-phase zone where separation of primary aluminides can occur during solidification. The aluminides are dense and will separate from the liquid without stirring and settle to the bottom of the casting during slow cooling. For alloys melted in a graphite crucible and poured into a graphite mold, it has been shown that as much as a 5% difference in primary aluminide concentration can exist for a 30 wt% U alloy.

The uranium-aluminum alloy system has three different intermetallic compounds, UAl_2 , UAl_3 , and UAl_4 . The composition of UAl_4 is about 67 wt% uranium and 33 wt% aluminum. The liquidus temperature for this composition is approximately 1500°C; also, a peritectic reaction occurs at a temperature of 731°C. The low temperature of the peritectic reaction requires a relatively long diffusion time at temperature for completion. Thus, the microstructure of a slow cooled casting most often consists of a mixture of both UAl_3 and UAl_4 intermetallic compounds. These compounds generally have different physical and chemical characteristics that differ from a solid solution alloy, which could prove beneficial for the corrosion resistance of the waste form.

The current recommended melt-dilute treatment process alloy composition is the uranium-aluminum eutectic with dilution to $\leq 20\%$ ^{235}U . This composition, 13.2 wt% U, has the lowest liquidus temperature on the U-Al Phase diagram. The melt and dilution process has been demonstrated for each of the three fuel types considered in the ATP at SRS. The relatively low temperature required to produce a pure liquid enhances the remote operability and safety of the

treatment process and limits the radionuclide species that will volatilize from the SNF during treatment.

5.2 Neutron Absorber Additions

Metallurgical compatibility of neutron absorber material additives to uranium-aluminum alloys defines the success of the fabrication process. Compatibility of these absorbers is indicated by the corrosion characteristics of the final melt-dilute product. The neutron absorber materials must not readily separate under corrosion degradation scenarios in order to maintain criticality control. The following sections discuss the drivers and development of the melt-dilute SNF form that incorporates neutron absorber materials.

The Al-SNF assemblies were originally fabricated using enriched uranium and aluminum alloy, with an aluminum alloy cladding. During reactor service, fission products are produced within the assemblies. The relatively small quantities of fission products by mass, however, are not expected to significantly alter the microstructure of the MD-SNF form. Uranium-aluminum alloys are used as surrogates to investigate the behavior and characteristics of actual MD-SNF. The alloys are made from surrogate fuel assemblies, melted and alloyed with additional aluminum, to obtain a near eutectic composition (~13.2 percent by weight U in Al). Neutron absorber materials are also added during the melt-dilute treatment.

The general microstructure of the uranium-aluminum alloy as the MD-SNF surrogate is that of a simple binary eutectic. The microstructure of the melt-dilute form without neutron absorber additions consists of UAl_x phases dispersed in an aluminum matrix. The eutectic composition is observed to be sensitive to cooling rate or impurity level, and hence, will not be the same under varying processing conditions [2]. If the melt composition deviates slightly from the eutectic composition, the microstructure will form primary Al or UAl_4 followed by the formation of a eutectic between regions of the primary phase. SEM and EDS analyses have also identified the presence of a primary UAl_3 phase in the near-eutectic uranium-aluminum alloy. The general microstructure is presented in Figure 5-2.

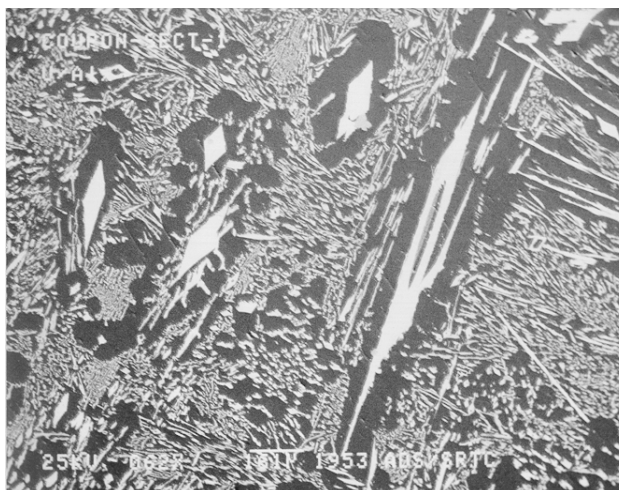


Figure 5-2 General Microstructure of the Melt-Dilute SNF Form

The addition of gadolinium and/or hafnium, in amounts up to a total of three percent by weight, to the MD-SNF form does not substantially alter the microstructure of the form. Near-eutectic

uranium aluminum alloys have been fabricated and evaluated through SEM and EDS analyses. These analyses indicate that the microstructures with and without the gadolinium addition are similar and that the gadolinium added to the melt-dilute form collocates with the uranium present in the UAl_4 phase.

The microstructure of the melt-dilute form with an addition of 1.5% gadolinium and 1.5% hafnium by weight is presented in Figure 5-3. The microstructure of this system is similar to that of the binary eutectic uranium-aluminum alloy shown in Figure 5-2 but is complicated slightly by the presence of additional hafnium-containing phases. SEM and EDS analyses indicate that the hafnium plus gadolinium addition to the melt-dilute form collocate with the uranium present in the primary uranium-aluminum phases within the SNF form.

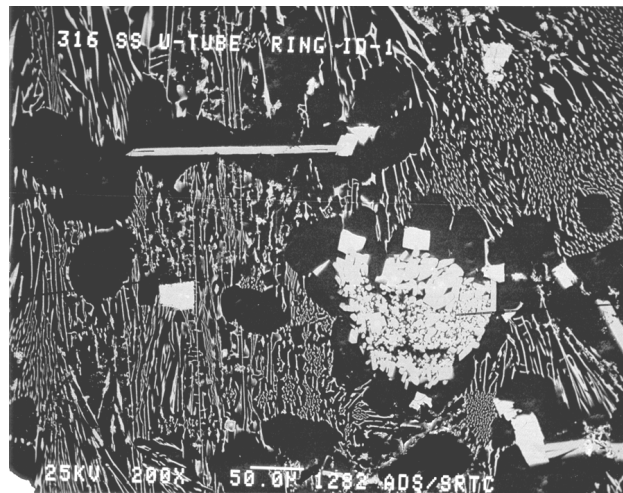


Figure 5-3. SEM Photomicrograph of the Surrogate MD-SNF Form with 1.5% Gd and 1.5% Hf by Weight.

The metallurgical evaluation of the surrogate melt-dilute/neutron absorber systems to date provides evidence that the melt-dilute treatment can integrate neutron-absorbing materials into the MD-SNF form uniformly throughout the ingot in U-Al phases. Both neutron-absorbing elements exhibit desirable alloying characteristics. Gadolinium additions tend to concentrate in the UAl_4 phase that is more corrosion resistant than the bulk MD-SNF form. Therefore, the gadolinium is expected to stay closely associated with fissile uranium in the UAl_4 phase as the MD-SNF form degrades, providing an additional barrier against the solubility facilitated removal of gadolinium from the waste package. Hafnium additions tend to concentrate with aluminum in an aluminum-hafnium phase or in a uranium-aluminum-gadolinium-hafnium phase. This makes the hafnium more susceptible to corrosion. However, the relative insolubility of hafnium would prevent it from being removed from the waste package. The combination of gadolinium and hafnium for criticality control provides the highest probability for maintaining criticality control of the MD-SNF form.

5.3 Pre-Treatment Drying

The melt-dilute process includes molten metal, making it imperative that the Al-SNF assemblies are dry. The introduction of liquid water into molten metal can result in a steam explosion. Therefore, the MD process includes pre-treatment drying of the SNF assemblies prior to treatment to preclude the possibility of melt-water interactions. Melt-water interactions or steam explosions have the potential to effect injuries, fatalities, or loss of equipment. As metal

containing surface water or water trapped in crevices, etc. is added to molten alloys, the free water instantaneously vaporizes with a subsequent volume expansion resulting in a steam explosion. With this steam explosion, molten metal is expelled rapidly from the crucible. Work at Oak Ridge National Laboratory during the 1960's showed that the violent nature of steam explosions increases with increasing melt temperature and water volume [3,4,5].

The drying stage within the MD process seeks to prevent or eliminate the possibility of a steam explosion by removing the free water from the SNF assemblies prior to contact with molten metal. Spent nuclear fuel assemblies that are stored in wet basin storage are removed from the basin and allowed to "drip" and air-dry in Lag storage prior to movement of the assemblies to the MD treatment room. This will result in only minimal free surface water for removal from crevices and voids in the assemblies with a drying operation.

Common practice within the commercial aluminum foundry industry is to use a drying oven to remove moisture from process crucibles, charge materials, and casting utensils (dip ladles, slag rakes, and thermocouples). These ovens are typically temperature limited ($< 400^{\circ}\text{C}$) and resistance heated. Some models use a heated forced air convection arrangement to remove moisture.

The drying oven should be designed to be large enough to hold the bundles of fuel that will be processed during one day. It must be designed such that it is inherently critically safe under all operating conditions. The oven will operate at a slight negative pressure with the offgas being vented to an exhaust system containing a HEPA filter that is designed to accept moist air that is possibly contaminated with fission gases, tritium, and/or particulate material.

The temperature of the oven containing the fuel or fuel bundle will be increased to 200°C and held at that temperature for a minimum time that has been demonstrated effective in the removal of all liquid water. This time is expected to be about two (2) hours. Dried fuel is transferred directly from the drying oven to the melt-dilute furnace.

5.4 Melt-Dilute Casting Techniques

The ATP at SRS investigated three casting methods, two of which, top pouring and bottom pouring, involve a separate crucible and solidification vessel and one in which the crucible is used as the actual solidification vessel, in-situ solidification. A top pour is a physically simple operation that merely requires rotating the crucible to allow the molten metal to pour into the mold or casting vessel. A bottom pour requires the use of a stopper rod or slide gate to regulate flow into the mold or casting vessel. In-situ solidification alleviates the problems inherent in pouring processes and simplifies the offgas system requirements due to the lack of a pouring step that will promote evolution of volatile radioactive species from the melt.

Solidification in crucible provides the slowest cooling rate and simplifies the microstructure of the SNF form. It is an inherently safe process because there is no chance of spillage. The lack of melt disturbance (as would be present after pouring) reduces the potential for porosity in the casting and reduces the amount of volatile species released to the offgas system. Solidification in crucible also allows the slag and oxides to become an integral part of the casting and eliminates the need for separate disposal. The recommended technique is therefore to solidify the melt in the crucible. This relatively simple method allows ease of offgas system and remote handling operations. The integrity of the offgas system will not have to be broken in order to accomplish a pouring operation. Additionally, since no molten metal stream will be passing through a pouring orifice, there is no chance for pour spout clogging/degradation. Proper design and selection of

the crucible materials should allow for (1) easy removal of the solidified ingot from the crucible or (2) placement of crucible and ingot in canister for storage, making it compatible with remote handling. The current melt-dilute treatment technology employs the disposable crucible liners that will be disposed with the melt-dilute SNF form, thereby decreasing secondary waste, crucible degradation, and process room contamination.

5.5 Crucible and Liner Materials Specification

The choice of the appropriate crucible materials depends on several variables including temperatures, melt interaction, and batch size. Several different crucible materials have been evaluated for use in the melt-dilute treatment process. These materials are graphite, carbon steel, and oxide ceramics, which are typical commercial foundry crucible materials. Work on the ATP at SRS has led to the selection of a graphite crucible with a carbon steel liner. The graphite crucible provides better process characteristics at the elevated temperatures required for the melt-dilute treatment process. The carbon steel liners are expected to interact with molten aluminum at the melt-dilute process temperatures approaching 1000 °C. However, experimental data indicate that the reaction between the carbon steel liner and the melt-dilute alloy is sufficiently sluggish that if the time at temperature is minimized, the relatively inexpensive carbon steel liners are suitable for the melt-dilute treatment process.

5.6 Batch Size

It is currently envisioned that the crucible for the melt-dilute treatment process should be able to accommodate 15-20 MTR type elements or a single HFIR core. The current design for the melt-dilute technology produces an SNF form that will fit the diameter of a canister with a 17 inch diameter that is 120 inches in length. Thus, in order to ensure that the melt-dilute ingot resulting from the treatment process is able to fit into the canister the designed crucible dimensions for the melt-dilute process are 16.5 inches in diameter by 36 inches in height. A crucible of this size would have a volume of approximately 135,000 cc and can easily accommodate 15-20 MTR type assemblies. Mechanical size-reduction or reduction in the number of assemblies per batch can accommodate oversized assemblies.

5.7 Furnace Requirements

Preliminary design and definition of the functional performance characteristics of the melt-dilute treatment process facility are focused on the operation of melting irradiated fuel assemblies. This section details the basic requirements for the melt furnace with respect to type, power, stirring, and potential operating mode.

5.7.1 Furnace Type

The two basic furnace types that have been evaluated during the development of this treatment technology are resistance and induction. Small-scale melting facilities have been designed and operated with both furnace types. Initial experiments conducted in the small-scale resistance furnace apparatus (Figure 5.4) showed several disadvantages to using this type of system and lead to the design and construction of the small-scale induction furnace apparatus used in our integrated process demonstration. Two of the disadvantages to using a resistance furnace system are related to melt stirring. Since, there is no built-in stirring capability with a resistance furnace an external mechanical stirrer is needed. This adds complexity to the apparatus as well as mechanical stirring being less efficient than induction stirring. Additionally, removal of the mechanical stirrer presents problem with respect to the spread of contamination. Thus, the recommended furnace for this operation is a standard commercial water cooled induction furnace with a power supply designed to couple with the selected crucible material that is required for this

facility. The frequency of the induction power supply will have to be determined by the furnace vendor but is likely to be in the range of 300-1,000 Hz.



Figure 5.4 Small Scale Resistance Furnace Apparatus

Modern induction furnaces operate at frequencies of 70-5000 Hz, allowing high power densities and producing a natural stirring action in the melt. There are two types of induction furnaces, channel and coreless.

The channel type furnace has a lower chamber that connects to a channel that wraps completely around the primary induction coil. This type of furnace has two inherent drawbacks. Only a small portion of the metal (that inside the channel) receives energy at any one time, limiting the energy transfer. A molten heel must be kept in the lower chamber and channel at all times. If the material completely solidifies, refractory damage will result and restart of the furnace will necessitate a small molten charge to initially fill the channel.

The coreless furnace, which is preferred for this application, uses the crucible and the metal inside as the secondary. The primary wraps completely around the crucible, in the form of a water-cooled copper coil. The advantage of this design is that it may be completely emptied, has high-energy transfer, and is simpler in construction. Another advantage is that the melt is stirred more vigorously because the field may couple with the entire molten volume.

5.7.1.1 Power and Cooling

A primary transformation from the high-voltage grid of the local utility is usually required because the voltage applied to the coil is between 500 and 3000 V. The typical conversion is in an ac-dc-ac sequence.

A closed-loop water system is generally used to cool the furnace coils and power supplies of the induction furnace. In a medium-frequency power supply, the electronic components of the system contain a dc leg; it is therefore important to have clean, demineralized water circulating, typically with conductivity set at 50 mS (50mmho) in order to prevent electrolysis from taking place.

5.7.1.2 Melting Operations

There are two ways to operate a coreless induction furnace. One involves completely emptying the furnace of molten metal each time and recharging it with solids for the next run. The other involves a tap-and-charge operation in which one-third to one-half of the furnace contents are emptied each time, leaving a molten heel in the furnace at all times. There are advantages and disadvantages to each method.

The first method has the advantage that the crucible may be used as the mold, eliminating some of the complexity of the melt-dilute process. There is also a power advantage because medium frequency power supplies produce a flux that couples with the cold charge material more effectively than it does with the molten metal. In addition, this method allows the drying of wet or oily charge material in the furnace, minimizing the need for a drying system. In order to avoid bridging, the length of the solid pieces in the charge should be no greater than two-thirds of the crucible diameter. Longer sections should be cut or sheared to length before insertion into the crucible. This is the preferred method for the melt-dilute process.

The second method has the advantage of producing longer refractory life due to reduced thermal cycling and improved melt chemistry because less of the furnace contents need to be replenished at one time.

5.7.2 *Stirring*

In addition to an induction power supply required to heat the crucible, an induction stirring power supply will also have to be provided for this facility. The stirring power supply will operate at 60 Hz.

When alternating current is applied to an induction coil, it produces a magnetic field, which in turn generates a current flow through the charge material, heating and finally melting it. The amount of energy absorbed by the charge depends on the magnetic field intensity, electrical resistivity of the charge and the operating frequency. The induced current in the charge creates a second magnetic field. Because these two fields are always in opposite directions, they create a mechanical force that is perpendicular to the lines of flux and cause metal movement or stirring when the charge is liquefied. The mechanical force stays perpendicular to the field only in the center of the coil; on both ends of the coil it changes direction. The metal is pushed away from the coil, moves upward and downward, and flows back. This is referred to as four-quadrant stirring. The stirring action aids in alloy and charge absorption and produces a more chemically and thermally homogeneous melt. The stirring is directly determined by the amount of power induced and is inversely proportional to the square root of the frequency. Therefore, increasing the power while lowering the frequency will result in increased stirring intensity. The two power supplies for the furnace and stirring unit are independent of each other and are designed so they can never operate simultaneously.

5.7.3 *Furnace Power*

The furnace induction power supply should be designed to deliver the proper level of power to the furnace in order to melt the expected charges of 15-20 MTR assemblies or a single HFIR

core. Current understanding of the operation of the melt-dilute treatment process facility indicates that a single melt batch will be run pr day. Thus, the furnace power supply should be able to melt the appropriate size charge and addition materials in a manner such that the entire melt batch procedure can be completed in a single operating shift. A power supply unit in the range of 200-300 kW would be fully capable of meeting these needs.

5.7.4 Atmosphere

The entire melt-dilute process will take place in an air atmosphere. No special cover gas is specified. The inside of the melt crucible will be at a slight negative pressure to minimize the possibility of escape of any evolved gases. In considering atmosphere, it should be noted that graphite at high temperature would react with air to form gaseous oxides of carbon. Thus, should graphite be the crucible material of choice the atmosphere above the melt is likely to be deficient in oxygen, and it may be necessary to supply air if available oxygen is required to react with gasses that are evolved from the melting fuel.

5.8 Process Cycle Options

Two casting processes were considered in the development of the melt-dilute technology. These are superheat casting or mushy-zone casting. The desire for uniformity in the melt-dilute SNF form led to the selection of the superheat casting process that involves heating the desired alloy to some prescribed temperature above the alloy liquidus temperature. Mushy-zone casting involves heating the alloy to a temperature below the liquidus within a two-phase region where one of the two phases is a liquid and the other is a solid and would not provide complete homogeneity in the melt-dilute SNF form.

A schematic of the treatment cycle for the melt-dilute process is shown in Figure 5.5. The basic schematic shows heating of the alloys above the liquid temperature but not to exceed 1000°C. Once at temperature, depleted uranium and aluminum is added to achieve the desired final alloy composition and process sampling is performed to monitor and control the treatment process.

For the binary U-Al fuels the desired final alloy composition as previously specified is the eutectic composition. The liquidus temperature for a eutectic alloy is 646°C. Thus, the treatment cycle temperature is in the range of 800-850°C to allow for proper melt homogenization. Once the highly enriched assemblies are melted, depleted uranium for dilution and aluminum for composition control are added to the melt. Sampling of the melt prior to and following the additions of the uranium and aluminum is performed for process control and verification. The remaining sampled material is used for post-treatment characterization. The casting is then cooled to room temperature, removed from the crucible, and transported to the packaging station. Processing time is expected to be less than 2.5 hours.

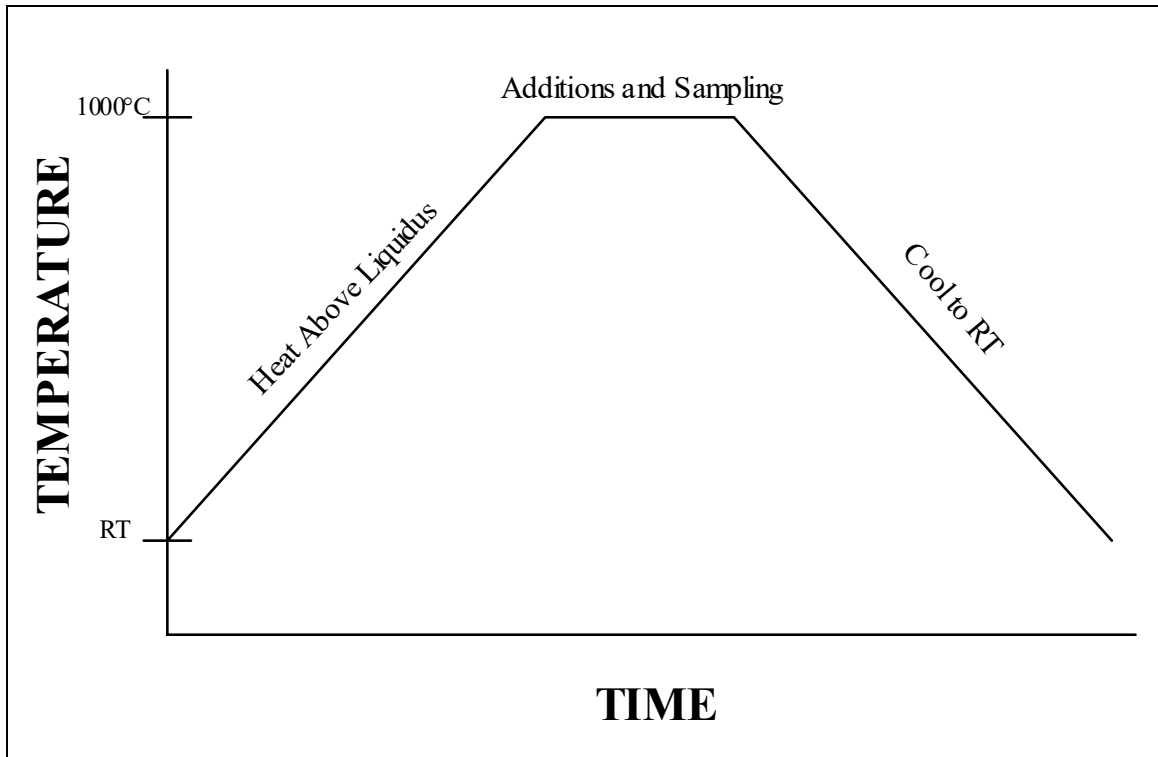


Figure 5.5 Schematic Treatment Cycle for the Melt-Dilute Treatment Process

The ternary isothermal section for a uranium-aluminum-silicon that has been developed in the ATP at SRS to determine the liquidus temperature, it is apparent that no treatment cycle temperature adaptations are necessary for U-Al-Si fuels. This has been confirmed in simulator testing as part of the ATP. Experiments and literature data on the melting of alloys consistent in content with the oxide fuels indicate that treatment of the oxide fuels to effect complete dissolution of the uranium oxide particles and allowing isotopic dilution of the enriched uranium can be achieved without any treatment cycle temperature adjustments. However, the time at temperature may have to be extended to assure complete reduction of the uranium oxide within the melt.

5.9 Development of Melt Treatment Process for Other Fuel Types

5.9.1 Aluminum-Clad, U Metal Fuel

The treatment of aluminum clad, uranium metal fuel is ideal using the melt-dilute technology. Treatment of this fuel type would not focus on the dilution of enrichment since these fuels were initially only 2% enriched, however, the focus would center on volume reduction for ultimate disposal. The Melt-Dilute technology provides approximately a 70% volume reduction by consolidating the fuel into a compact ingot. This leads to fewer waste disposal canisters and ultimately lower overall disposal costs. Alloying the uranium metal fuels with aluminum will be similar to treating the U-Al alloy fuels which have been discussed in detail in the previous sections.

5.9.2 Aluminum-Clad, U_3O_8 or UO_2

For the Melt-Dilute treatment of the uranium-oxide powder metallurgy fuels, the uranium-oxide reacts with the aluminum metal to form a U-Al alloy and a slag of aluminum oxide (Al_2O_3) floating on top of the melt. However, the chemical kinetics of oxide conversion in aluminum are slow. By the addition of elements such as calcium, the oxides are readily reduced and dissolution becomes rapid. The calcium is not a catalyst, but is a process chemical that must be added in sufficient quantity to reduce the heavy metal oxides. The slag in this case is predominantly calcium oxide (CaO) with whatever metal attaches itself to the slag. With the complete reduction of the uranium-oxide by the calcium addition, the uranium metal and aluminum will alloy to form a uranium-aluminum alloy.

5.9.3 UO_2 with Mg Fuel

The treatment of UO_2 -Mg powder metallurgy fuels using the Melt-Dilute technology would require a significant research effort. Several issues are raised with treating these fuels using this technology—two of the more prominent are 1) handling/melting issues with uranium metal, and 2) volatilization of magnesium. Magnesium is a highly reactive metal. The alloying of aluminum and magnesium is not a trivial process, as great care must be taken to avoid ignition of the magnesium. Additionally, magnesium possesses a relatively low vapor pressure and in many cases is alloyed under pressure with other metals to avoid extensively volatilization. Excessive volatilization of MgO from the surface of the melt-dilute melt could result in many challenges that the current off-gas system is not designed to handle.

5.9.4 Stainless Steel-Clad, U-Cr-Ni Fuel

The treatment of stainless steel clad, U-Cr-Ni fuel would require moderate research and development. The melt-dilute treatment technology has previously shown that stainless cladding/canisters can be readily dissolved by aluminum. The only issue posed by these fuels is that of the behavior of the fuel meat. If the nickel and chromium alloying level in the uranium are low it is anticipated that these materials would behave similarly to the U-metal fuel which readily alloys with aluminum. However, should the nickel and chromium additions be significant a moderate research effort would be needed to evaluate their effect on the processing temperature and kinetics.

5.9.5 Zirconium-Clad, Uranium-Zirconium Fuel

The Melt-Dilute technology using aluminum to treat Zr-clad, U-Zr fuels is not attractive. The phase diagram for the Al-Zr system shows a steep liquidus slope for high Al concentration alloys. This steep liquidus translates into increasing processing temperatures in order to alloy significant quantities of Zr with Al. Thus, with increasing processing temperatures other challenges such as increased off-gas arise. To keep the processing temperature similar to that used for the other fuels, large amounts of Al can be used, however, this will result in a waste volume increase rather than a decrease. With these issues in mind, it is not recommended that Zr-clad, U-Zr fuels be treated using the Melt-Dilute technology.

5.10 Fission Product Volatilization

A large number of radionuclides with widely varying release characteristics exist in the reactor core at the end of an operating cycle. The elemental boiling points of these radionuclides are used as the delimiting characteristic for estimating the magnitude of release, because the primary release mechanism is vaporization of volatile species. Once the boiling point of a species is surpassed, the species will begin to evolve from the molten pool. If the molten pool remains at a given temperature for a long period of time, elements with boiling points below this temperature

will eventually be completely released. Additionally, examination of the vapor pressures of the radionuclide species indicates that some level of volatilization can occur at temperatures below the boiling point of the element.

Based on the data accumulated from the review of the literature and information on elemental melting point, boiling point, half-lives, and vapor pressure, a listing of the potential radionuclide species that may volatilize at treatment process temperatures in the range of 850-1000°C is shown in Table 5.1 below.

Table 5.1 Elements with Boiling Points Below 1000°C

Element	Symbol	BP (°C)
Tritium	^3H	-248
Krypton	^{85}Kr	-153.22
Selenium	^{79}Se	685
Xenon	^{133}Xe	-108.04
Iodine	$^{129,131}\text{I}$	184.4
Cesium	$^{134,135,137}\text{Cs}$	671
Rubidium	^{137}Rb	688
Tellurium	^{90}Te	988

This list is representative of radionuclides expected, but may not be totally complete as some of the higher boiling point elements may associate with some of the more volatile species and be released at the lower temperatures. Barium (1140°C) and Strontium (1382°C) have been seen coming off above 1050°C in experiments conducted at the Argonne National Laboratory. The main radionuclide species of interest is ^{137}Cs due to its relatively high concentration and effective dose. Volatilization of cesium begins at approximately 660°C. According to the literature the fraction of the cesium that can be expected to volatilize can vary from 5-80% depending on temperature and atmosphere. Effective capture of cesium has been demonstrated at temperature from 500°C and below.

Control of the atmosphere within the offgas system is crucial to ensure proper flow of the offgas stream through the filter media. The only two major requirements of the atmosphere are that 1) the atmosphere provided must be able to channel the volatilized radionuclide through the offgas media so as to effect efficient capture of the species, and 2) the system will need to be operated with a slight negative pressure to prevent release from leaks at seals. This will ensure an influx of atmosphere from outside the offgas system and prevent an escape of treatment process gas to the atmosphere.

5.10.1 Filter Media

Any furnace design will require an offgas system to capture volatile fission products. The primary contributor to offgas radioactivity will be ^{137}Cs with smaller amounts of ^{85}Kr , ^{99}Tc , and ^{129}I . This section describes some of the available options for design and operation of the offgas system. All the methods considered include some means for condensing the Cs and Tc followed by a HEPA filter to remove particulates. An iodine absorption bed may also be required. We are assuming for the present that the Kr will be released to the atmosphere. If it must be trapped, a silver mordenite bed operated at dry ice temperature will be used. Several means for final disposition of fission products removed from the offgas are being studied.

5.10.1.1 Cesium Condenser

The simplest method for removal of cesium from the offgas stream is to cool it to a temperature below its boiling point and to trap the resulting condensate. This will likely be accomplished by use of a metallic labyrinth or other medium on which cesium can be cooled and condensed. The cesium must be kept above its boiling point until it reaches the condenser to prevent its plating out on the surface of the furnace and offgas piping. Possible media for effecting capture of the cesium are glass frit, zeolite, activated alumina, activated charcoal, and crystalline silico-titanate.

Cesium metal melts at 28.5°C and boils at 670°C. The simplest and most effective method for removal of Cs from a hot gas stream is to cool the gas to a temperature well below cesium's boiling point, condensing out the Cs on a cool surface. The Cs can then be disposed of on the condenser medium or be dissolved in an aqueous wash that can then be disposed of to the HLW tanks. The condenser can be located anywhere that its temperature can be kept below 670°C. The most likely sites are the furnace lid or a trap immediately adjacent to the furnace.

Bench-scale laboratory scoping studies were conducted using non-radioactive cesium. The outcome of these tests indicated that two absorber bed media, activated alumina and zeolite 4A, were acceptable for cesium trapping. Following these tests a full-scale integrated melting and off gas apparatus was constructed. This apparatus was capable of melting a single full-size (3" x 3" x 36") surrogate fuel assembly. Three different absorber bed designs using zeolite 4A as the trapping media were evaluated. The goal with each design concept was to completely capture cesium vapor by maximizing residence time and velocity. A design concept termed the concentric ring design produced the most consistent result with a nominal system efficiency of 99.9%. Further melt-dilute tests using irradiated SNF coupons were also performed at Argonne National Laboratory in 2000. The results from these tests validated the analytical modeling and laboratory studies. The outcome of these experiments was two-fold in that other than the noble gases and iodine only Cs-137 was shown to volatilize from the melt and a zeolite 4A absorber bed allowed zero breakthrough of cesium during testing.

The analytical and experimental tests conducted at both SRTC and ANL, has identified cesium as the melt constituent of most concern with respect to volatilization. Experimental tests using both cesium surrogates and radioactive cesium have shown that zeolite 4A is an effective cesium trap and as a result a preliminary offgas system concept has been developed employing dry zeolite 4A absorber beds as the primary cesium trapping medium.

Based on the outcome of the analytical modeling that cesium is the major concern an experimental program was begun to develop an offgas system to effect the capture of cesium from the off-gas stream.

5.10.1.2 Iodine Reactor (if needed)

The SRS H-canyon aqueous process utilizes an iodine reactor consisting of a vessel filled with berl saddles on which silver nitrate has been coated. This reactor is maintained at a temperature between 180 and 200°C. Other designs for iodine reactors have been considered; most incorporating a silver-loaded, heated bed. An activated carbon bed is also effective for I₂ removal. Although any one of these methods could be utilized in the melt-dilute offgas treatment process, the activated carbon bed will probably provide the least expensive option if a temperature of less than about 130°C can be maintained; otherwise a silver-loaded reactor will have to be used.

5.10.1.3 Krypton Absorber (if needed)

The issue of krypton absorption is primarily a regulatory one. Preliminary calculations have shown that the radiation dose at the site boundary from the release of this krypton will not exceed 0.04 mrem/year [6], significantly less than the maximum 3 mrem/year dose permitted by 10CFR50, Appendix I. Therefore, for planning purposes it is assumed that any Kr released will be vented to a stack without any attempt at removing it from the offgas stream. Should it become necessary to remove the Kr, a silver mordenite or hydrogen mordenite bed, operated at dry ice temperature, will be effective [7]. The primary difficulty in using the Kr absorber is the necessity to operate it at dry ice temperature with a gas stream originating in a 1000°C or hotter furnace.

5.10.1.4 HEPA Filter

No matter what other treatment processes are used to remove fission products from the offgas stream, a HEPA filter will be used to remove any remaining particulate matter before the gas is finally released to the atmosphere via a stack. HEPA filters are available that use either a replaceable glass medium or a permanent (cleanable) stainless steel medium.

5.10.2 *Flow Rates and System Pressure*

The melter system will operate at a slightly negative pressure relative to the atmosphere. The airflow in the bench-scale melter was adjusted to provide approximately three to four air changes per minute (approximately 3.5 scfh). Using the same criterion, the pilot-scale melter will be operated at 3.5 to 5 scfm. To obtain this airflow rate, the initial experiments in the pilot-scale melter will be operated at approximately 5 inches water column. It is expected that the operating pressure in the final system will be in the range of 3-5 inches water column.

5.10.3 *Monitoring Filter Capacity*

In order to ensure effective capture of the volatilized radionuclides and prevent blow-through it is necessary to monitor the capacity of the filter beds. Additionally, safe storage, handling, and transportation of spent filter media are important and as such monitoring activity levels in these media is paramount. Thus, the offgas should be able to monitor the activity levels within the filter beds and offgas media in order to ensure proper operation of the system and also to allow for easy cost effective handling, transportation and disposition of the spent offgas media.

The primary radionuclide of concern is Cs-137. Because it is a gamma emitter, it will be easy to monitor the accumulation of cesium in the sorber material by means of a gamma monitor or ion chamber placed near the filter chamber and shielded from the material in the furnace.

5.10.4 *Stack Emissions*

Stack emissions of I-129, Kr-85, and tritium are predicted to be well below plant release guides and are therefore not considered here. Bench-scale experiments with cesium and aluminum have shown that the cesium released from the melt is less than 2.5% of that charged to the melter. This can be reduced further by the addition of certain materials directly into the melter that trap and immobilize the cesium. The cesium released from the melt will be efficiently trapped on either a zeolite or activated alumina bed. In the bench-scale experiments, essentially no cesium has gotten past the adsorber beds. A HEPA filter will be located downstream of the melter offgas system to catch any particulate material that might get through the sorber bed so that stack emissions will be of little consequence.

5.11 Disposition of Secondary Waste

Selection and design of a system for scrubbing and disposing of cesium from the offgas for the Melt/Dilute operation is expected to be simple and straight forward for the following three reasons. (1) Only a small fraction, < 5%, of the total cesium in the Melt/Dilute operation is transferred to the offgas. Thus, the mass of cesium in the offgas is small. (2) Either a dry or a wet method can scrub cesium from the gas easily and efficiently. (3) There are several simple and practical methods available for final disposal of the cesium. Figure 5.6 presents the alternate methods, which have been identified for scrubbing and disposing of cesium in the offgas.

Note that the methods divide into two categories: dry or wet scrubbing. The dry scrubbing methods as depicted in Figure 5.7 are preferred and recommended on the basis of fewer process steps and a smaller and simpler HEPA subsystem, since the gas flow is smaller and hot and dry. The dry scrubbing methods must be further evaluated to develop the basis for final selection. The evaluation will include characteristics such as regulatory requirements/permissibility, capital and operating cost estimates, acceptability to the operating division, and acceptability to the organization receiving the cesium.

The simplest system for scrubbing and disposing of the offgas cesium merely places the dry cesium sorbant in the same canister with the melted, diluted fuel rods. The steps of this process are indicated in Figure 5.6. The cesium sorbant, a dry granular material, is held in an aluminum container. The cesium is captured by the sorbant as the offgas flows through it. When the sorbant becomes loaded, the container is removed and added to the storage canister. The offgas will pass through a stack of several sorbant containers so that the bottom container can become fully loaded while the upper containers prevent cesium breakthrough. A fresh sorbant container is added to the top of the stack as a loaded container is removed from the bottom. If necessary, the dry cesium sorbant could be placed in a separate canister by itself.

Preliminary shielding calculations for the handling and transportation of the cesium-loaded sorbant via one of the dry methods have been performed. The shielding required to reduce the dose rate at the side of the sorbant canister to below 200 mrem/hr is between 4.5 and 6.75 inches of lead depending on the source (i.e., only cesium or all of the isotopes).

Although the dry scrubbing methods have been selected as the preferred options, several wet methods are also available. The wet methods typically involve a leaching step that results in a high level liquid waste and potentially a low-level solid waste. However, these methods are not preferred because of their additional complexity/processing steps and development requirements.

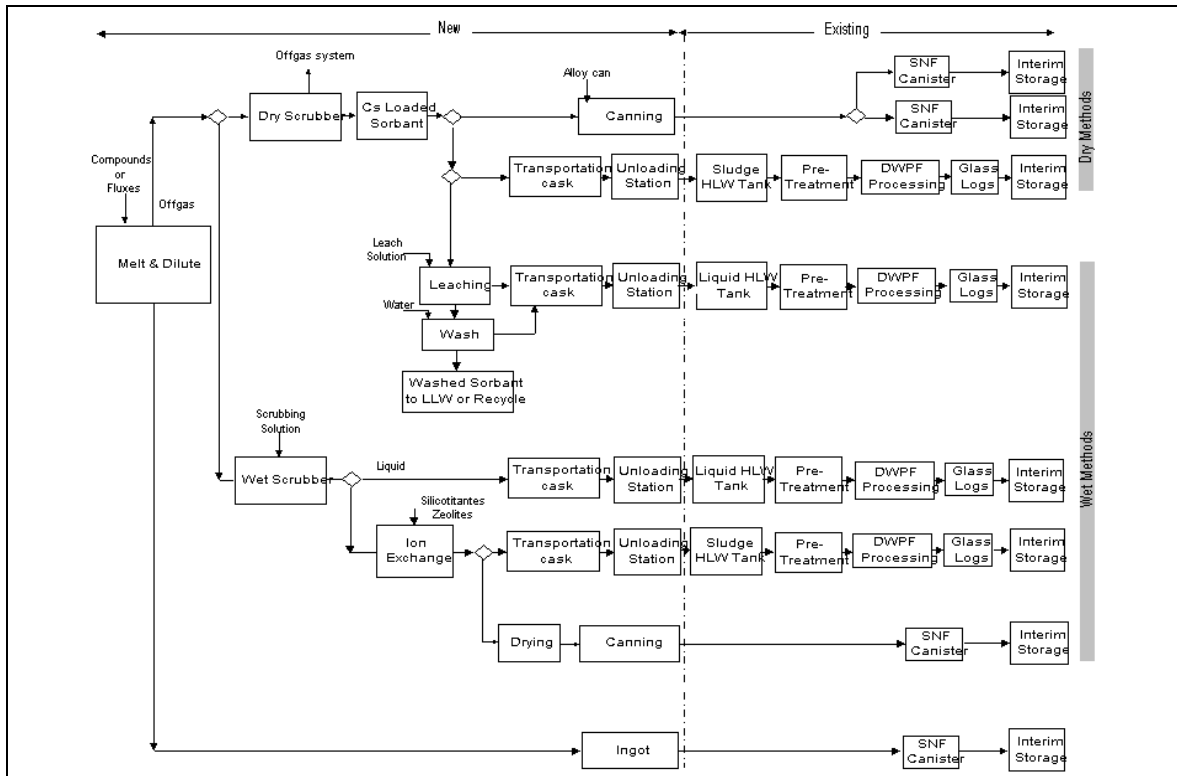


Figure 5.6 **Flow Sheet of Melt-Dilute Waste Disposal Treatment Options**

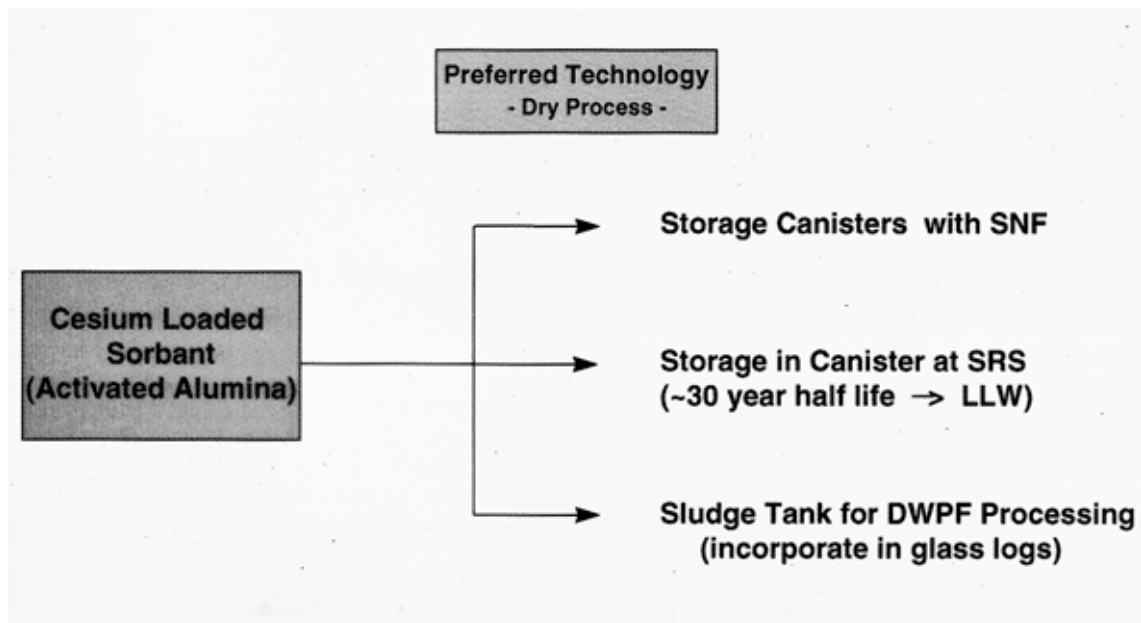


Figure 5.7 Dry Option for Cesium Loaded Filter Bed

6.0 STORAGE/DISPOSAL CANISTERS

Considerations for the preparation and packaging of melt-dilute (MD) waste forms for long-term storage or final disposal include identification and quantification of materials used as neutron absorber materials, backfill gas selection, canister design, and canister labeling. The topics discussed in the following two subsections are aligned with the requirements for interim dry storage at SRS and U.S. geologic repository disposal. The case considered is for the melt-dilute ingot that would be loaded into a special canister for long-term storage or disposal following the production of the ingot.

6.1 Packaging

6.1.1 Identification and Quantification of Materials Used as Neutron Absorbers

Analysis of the melt-dilute form has been performed using the methodology [8] approved by the U.S. Nuclear Regulatory Commission for U.S. repository disposal [9]. At 20% enrichment, minor amounts of n-absorbers must be added to meet the criticality requirements of $k_{eff} < 0.95$ for all mechanistically credible degradation scenarios.

6.1.2 Backfill Gas

The facility must maintain the capability of adding helium gas backfill to the DOE SNF canister [10]. The canister must be evacuated after loading, backfilled with helium to 1.5 atmospheres, weld-sealed, and leak tested.

6.1.3 Canister Design

The preliminary DOE SNF canister design/construction specifications provided in the recent WASRD [10] are summarized herein. The materials of construction for the DOE SNF canister shell and lids shall be selected to be compatible with the waste disposal container and with the contained SNF. Canister materials shall not corrode or otherwise chemically attack the waste package from the inside, and they shall not increase rates of degradation of contained SNF or mobilize radionuclides for transport subsequent to waste package breach. This material will be a low carbon austenitic stainless steel. The shield plug will be constructed of depleted uranium, sheathed in a compatible material such as stainless steel or carbon steel, or other high density materials except lead, without sheathing if compatible with the basket and shell materials.

At the time of acceptance into the CRWMS, disposable multi-element canisters shall be capable of sustaining a flat bottom drop from a height of 23 feet and a drop in any orientation from a height of 2 feet (individually – not both in sequence) onto an essentially unyielding surface without releasing radioactivity exceeding limits shown in Table 6.1 [10].

Table 6.1 DOE SNF Canister Release Source Term Criteria

Canister Type	Canister Release Dose-Equivalent Source Term Rem/Canister	
	Effective [TEDE _{canister}]	Max Organ [(CDE+DDE) _{canister}]
DOE SNF 18 in. diameter canister	1.15E+08	1.15E+09
DOE SNF 24 in. diameter canister	1.38E+08	1.38E+09
MCO	1.73E+08	1.73E+09

DOE SNF canisters shall be designed to support their own weight and that of their contents for multiple vertical lifts and horizontal translations while suspended from above via their lifting features.

6.1.4 Canister Labeling

The facility must maintain the capability of labeling each DOE SNF canister [10] with a unique, permanent alphanumeric identifier. The identifier will be unique to the waste-generator. It shall be visible from the top and side of the canister. It shall be in the form of a label that is an integral part of the canister, can be reasonably expected to remain legible for 100 years at temperatures of 25-400°C, does not impair the integrity of the canister, and is chemically compatible with the canister. Multi-element disposable canisters shall also have an exterior marking to indicate the SNF form contained within the canister.

6.2 Road-Ready Storage

6.2.1 Interim Dry Storage Requirements

Requirements to limit degradation of Al-SNF throughout the process of receipt, handling, drying, and interim storage are provided in Reference 11. Additional requirements were imposed to provide a road-ready canister and these are contained in reference 12. For example, the requirement for a helium gas backfill was added to facilitate leak detection in the canisters, to provide improved heat transfer characteristics of the filled canister, consistent with reference 10 requirements. Reference 13 demonstrates that these requirements are met for both the melt-dilute and the direct disposal processes as described in this report.

The site requirements for interim dry storage [11] are based on providing for safe, retrievable storage. Retrievability is directly related to limiting degradation in a storage system. In developing the SRS requirements for interim dry storage, limited fuel degradation was acceptable, consistent with the 10 CFR 72 requirement of no gross rupture of fuel cladding during storage and post-storage handling since the fuel was assumed to be eventually removed from interim storage. These criteria were developed prior to the decision to store MD form road-ready at SRS.

The application of “retrievability” for the road-ready storage of Al-SNF forms at SRS is adopted from SRS criteria for interim dry. This achieves both a safety benefit and a potential cost-benefit. The safety benefit includes affording safe conditions for postulated events in which the canisters may have failed and the MD form would need to be removed and re-packaged. If repository acceptance would necessitate a direct verification of Al-SNF condition, etc., to meet disposal criteria, a cost benefit is derived from the avoidance of opening canisters following storage at SRS (up to several decades). This is because the characterization information on fuel dimensions, physical condition, etc. assembled before canister sealing, would still be valid at the time of shipment.

Details on the potential degradation of aluminum cladding alloys and fuel materials in drying and dry-storage conditions are provided in References 11 and 14.

Temperature of the fuel must be below 200°C throughout storage in the TSF. This requirement is directly adopted from requirement for direct Al-SNF assemblies [11]. It is recognized that the

ingot produced in the melt-dilute treatment does not have the potential creep damage and hydrogen blistering that the direct treatment has and therefore this criterion is conservative.

6.2.2 Monitoring During Canister Storage

It is recommended that the ambient temperature and relative humidity external to the canisters be monitored throughout the dry storage period.

It is recommended that one or both of the following monitoring functions be performed for the canisters throughout the dry storage period:

- Provide for a lead surveillance canister that would be instrumented to measure internal canister environmental and AI-SNF form conditions for the typical storage configuration.
- Implement an inspection program using NDE to evaluate canister and AI-SNF integrity.

7.0 MELT-DILUTE SPENT NUCLEAR FUEL FORM ANALYSES FOR DISPOSAL SYSTEMS

The following sections, provided for reference, summarize analyses performed for the melt-dilute form for aluminum-based fuels to meet U.S. repository requirements. A complete listing of the requirements for U.S. Department of Energy fuel, and the demonstration of compliance to the requirements for the melt-dilute form, is provided in reference 15.

7.1 Melt-Dilute Characterization

The melt-dilute process allows for the direct in-process measurement of ingot composition through experimental, analytical, or computational means to verify product consistency and quality against the target composition. This measurement significantly reduces the need for pre-treatment characterization while significantly increasing confidence in final waste form characterization. The SNF form is significantly altered in the process and is inherently independent of the fuel fabrication and operation history.

For the melt-dilution process, the key parameters for process control are the total uranium content and the enrichment, which are used as input for melt composition calculations to determine the relative amounts of depleted uranium, aluminum, and/or poisons to be added in order to achieve the final target composition. These parameters may be determined from review of available fuel historical data or may be determined through analytical measurement in the melt during processing where irradiation and storage information is not available or is incomplete. Determination of these parameters during actual processing may be preferred in order to obtain a more accurate value for the homogeneous form. Thus, the melt-dilution process essentially eliminates the need to know the in-depth history of the fuel but would require an in-melt characterization to allow the most accurate characterization of the content. Highly precise numbers concerning total fissile species can be determined during the post-treatment characterization stage if deemed necessary. Unlike, the process control sampling step where time of analysis is crucial—need for quick, accurate response—there is no practical time constraint for the post-characterization step. During this characterization stage, the emphasis is on data precision and quality since it will ultimately be provided for further inputs regarding thermal analysis, dose, and performance assessment. Several commercial analytic techniques are available for providing these high precision values such as, gamma-ray spectroscopy, thermal ionization mass spectroscopy, and x-ray fluorescence spectrometry. Additionally, the possibility exist for the ability to perform bounding measurements and calculations such that each melt-grab sample need not be subjected to extensive analytic scrutiny in order to provide quality data for the variety of repository inputs. Upon completion of the dip sample analysis, the sample will be incorporated in the next melt batch.

7.2 Physical and Metallurgical Characteristics

The melt-dilute form for aluminum-based fuel consists of a casting from directly melting aluminum-based spent fuel assemblies, and adding depleted uranium to reduce enrichment to below 20% ^{235}U . The form is an ingot of a eutectic alloy (13.2%U in Al) that is comprised of an aluminum matrix with UAl_x phases.

The uranium is nearly all contained in a UAl_4 phase. Minor amounts of neutron-absorbing materials such as gadolinium and/or hafnium are also added for criticality control in amounts up to a total of 3 weight percent. Gadolinium additions tend to concentrate in the UAl_4 phase. Hafnium additions tend to concentrate with aluminum in a $(\text{U}, \text{Hf})\text{Al}_3$ phase with or without trace

quantities of gadolinium. Thus, the gadolinium is expected to stay closely associated with fissile uranium in the UAl_4 phase as the MD-SNF form degrades over geologic time in the repository, providing an additional barrier against the solubility facilitated removal of gadolinium from a degraded waste package.

7.3 Corrosion/Dissolution of Melt-Dilute SNF Form

Dissolution rates of radionuclides from both irradiated Al-SNF and unirradiated UAl alloys were measured under repository-relevant conditions including dissolution in water representative of that from the J-13 well at Yucca Mountain. The irradiated fuels had dissolution rates at 25 °C that ranged from approximately 0.2 mgU/m²/day (mgU/m²/d) for all the fuels in nominal J-13 to 30-100 mgU/m²/d in a nitric acid solution. Dissolution rates for radionuclides such as Cs, Sr, and Pu were approximately the same as the U dissolution rate over the duration of the test. The unirradiated alloys had dissolution rates ranging from 0.2 mgU/m²/d in nominal J-13 at 25 °C to 200-400 mgU/m²/d in low pH J-13 at 90 °C.

7.4 Criticality Analysis of the Melt-Dilute SNF Form

Neutron absorbing materials are needed to maintain $k_{eff} < 0.95$ in desired packaging configurations of MD-SNF with 20% ²³⁵U enrichment loaded in DOE-SNF canisters. Criticality of the MD-SNF loaded in a fully flooded, intact waste package with intact and degraded DOE-SNF canisters has been evaluated. Results of these preliminary analyses indicate that the melt-dilute SNF form requires the addition of neutron absorbing material to maintain criticality control. Candidate melt-dilute/neutron absorber systems include melt-dilute plus gadolinium, melt-dilute plus hafnium, and melt-dilute plus gadolinium and hafnium. These systems have been selected based upon thermal neutron absorption cross-section and upon geochemical considerations. The specific compositions have not been determined; however it is anticipated that less than 1% by weight of neutron-absorbing species in the melt-dilute form will be required to maintain reactivity limits.

7.5 Codisposal Waste Package Criticality Analysis

A criticality analysis was performed using the NRC-approved methodology in Reference 8. The worst-case bias, calculated from software code MCNP simulations of experiments described in Reference 9 includes a bias in the method of calculation and the uncertainty in the experiments. The interim critical limit of 0.93 was applied to the WP criticality calculations.

Three-dimensional Monte Carlo criticality calculations, for all anticipated intact- and degraded-mode configurations using DOE specified degradation scenarios, show that the requirement of $k_{eff} + 2\sigma$ values less than or equal to the interim critical limit of 0.93 is satisfied for the MD codisposal package if at least 7.5% of the original Gd loading (394.2 g) remains mixed with the fissile material. In the alternate MD ingot composition, Hf remains in the DOE SNF canister or waste package under all degraded conditions, thereby preventing a critical condition even if all Gd is removed from the degraded system.

Conversely, the distribution of hafnium makes its release from the waste package more heterogeneous than Gd and may make the hafnium more susceptible to release from the MD-SNF form by corrosion. However, the relative insolubility of hafnium (as compared to Gd, U and Al) should prevent it from being removed from the waste package prematurely.

7.6 Decay Heat Estimates

The development of the source term for decay heat of aluminum-based research reactor spent fuel assemblies per single assembly and the melt-dilute form per DOE canister has been completed. Bounding and nominal design source term values for use in thermal analyses of waste packages are developed. Decay heat calculations estimate that a DOE canister containing the melt-dilute form will have a heat load of less than 3,500 thermal watts.

7.7 Codisposal Waste Package Thermal Analysis

The engineering viability of disposal of spent nuclear fuel in a geologic repository requires a thermal analysis to provide the expected temperature history of the fuel waste forms within the disposal package. Calculated temperatures are used to demonstrate compliance with criteria for waste acceptance into the geologic repository and also to assess the chemical and physical behavior of the waste form within the codisposal waste package.

Thermal analyses of a codisposal waste package show that a helium-filled codisposal WP containing one AI-SNF canister and five Defense High-Level Waste (DHLW) glass logs having a 16 years cooling time, can satisfy a thermal design criteria for MD/SNF peak temperature criterion, $T_{\max} \leq 350$ °C, under the reference boundary conditions. In addition, these analyses show that average temperature of the WP decays close to geologic ambient temperature at about 2000 years of storage time after emplacement in a repository drift tunnel.

7.8 Waste Package Shielding Analysis

Shielding calculations for the proposed codisposal waste package estimate a maximum surface level dose level of less than 200 rem/h. In addition this value is well below a 10^4 rad/h value which has been shown to represent the onset of corrosion of materials used in the fabrication of waste packages when subjected to environments expected at the Yucca Mountain site [16].

8.0 ESTIMATED COST OF A MODULAR MELT-DILUTE FACILITY

The projected cost for a modular melt-dilute facility is \$10-20M. This figure includes design and fabrication of the modular facility and is based upon the experience gained through design and fabrication of the L-Area experimental facility at SRS.

9.0 ACKNOWLEDGEMENTS

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Appendix A

Table I

Russian Fuel Data

Facility Information			Fuel Assembly Data					Spent Fuel Data									
Country	Facility		Power MW	Critical Date	Shut Date	Fuel Assembly Type	Fuel Meat Type	Clad	Enr. %	Grams U-235 per FA	FA in Core	Average Discharge BU, %	No. Spent Fuel	Total kg U	Total kg U-235	Total Weight of FA	
	Name	Status															
U ₃ O ₈ -Al																	
Argentina												41	5	5.1	1	26	
Buenos Aires	RA-3	O	5	1968	-	MTR	U3O8-Al	Al	19.8	200	30		Converted to Russian LEU in 1990				
Germany																	
Dresden	RFR	S	0	1962	1991	Rods	U3O8-Al	Al	19.9	5	0		1,962	49.2	9.8		
Egypt																	
	ETR-2	O	22	1997	-	MTR	U3O8-Al	Al	19.8								
Totals														1967	54.3	10.8	
U-Al Alloy																	
Bulgaria																	
Sofia	IRT-Sophia	S	2	1961	1999	C-36	U-Al Alloy	Al	36.6	156	0		20	17	7.2	2.7	
Czech. Rep.	LWR-15					IRT-2M	U-Al Alloy	Al	80	171	0		32	235	50.2	40.2	
Rez	Rez	O	10	1957	-	IRT-2M	U-Al Alloy	Al	80	147	0		Ready to ship spent fuel to Mayak				
Hungary																	
Budapest	Reactor	O	10	1959	-	WWR-S M/M	U-Al Alloy	Al	36	39	227		50	1,112	120.5	43.4	
DPR Korea						IRT-2M	U-Al Alloy	Al	80	171				87	186	14.9	
Pyongyang	IRT-DPRK	O	8	1965	-	IRT-2M	U-Al Alloy	Al	80	147			3-tube (147 g U235), 4-tube (171 g U235)				284
Kazakhstan	WWR-K					WWR-C	U-Al Alloy	Al	36	110	38		35	312	96.3	34.3	
Almaty	Alm	O	6	1967	-	WWR-C	U-Al Alloy	Al	36	64	6		35	20	4.7	1.7	
Kazakhstan																60	
	IGR	O	10	1961	-	Hex Tubes	U-Al Alloy	Al	36		76						
Latvia						IRT-2M	U-Al Alloy	Al	90	149	0		30	10	1.7	1.5	
Riga	IRT-2M	S	5	1961	1998	IRT-2M	U-Al Alloy	Al	90	171	0		35	18	3.4	3.1	
Libya						IRT-2M	U-Al Alloy	Al	80	171	16-x		0	0	0	0	
Tajoura	JRT-1	O	10	1981	-	IRT-2M	U-Al Alloy	Al	80	147	x		0	0	0	0	
Poland																	
Otwock	EWA	S	10	1958	1995	WWR-SM	U-Al Alloy	Al	36	36.9	0		40	2,095	226.4	81.6	
Poland																1,896	
Otwock	AGATA	S	0	1973	1995	Multi-Tubes	U-Al Alloy	Al	80		0						

Facility Information				Fuel Assembly Data							Spent Fuel Data					
Country	Facility		Power MW	Critical Date	Shut Date	Fuel Assembly Type	Fuel Meat Type	Clad	Enr. %	Grams U-235 per FA	FA in Core	Average Discharge BU, %	No. Spent Fuel	Total kg U	Total kg U-235	Total Weight of FA
	Name	Status														
Poland																
Romania	Otrock	MARIA	O	1974	-	Multi-Tubes	U-Al Alloy	Al	80	345	6	35	275	118.5	94.9	1,774
	Bucharest	WR-S	S	1957	1997	C-36	U-Al Alloy	Al	36	141.7	0	39	70	27.6	9.9	210
Ukraine	Kiev	WWR-M	O	1960	-	WWR-M2	U-Al Alloy	Al	38	37	91	50	247	25.4	9.1	222
Uzbekistan						IRT-3M	U-Al Alloy	Al	90	270	0	45	213	63.9	57.5	788
Tashkent		WWR-CM	O	1959	-	IRT-3M	U-Al Alloy	Al	90	300	0					
						C-36	U-Al Alloy	Al	36	141.7	0		2	0.8	0.3	6
Viet Nam		Dafat										12	0	0	0	0
Dalat		Resea	O	1963	-	WWR-2M	U-Al Alloy	Al	36	40.2	100		No spent fuel until the end of 2000			

UO₂-Mg

Bulgaria	Sofia	IRT-Sophia	S	2	1961	1999	EK-10	UO ₂ -Mg	Al	10	128	0	30	58	74.2	7.4	203
Belarus	Minsk	IRT-M	S	4	1962	1988	Spheres	UO ₂ -Mg	Al	10	128	0	20	34	43.5	4.4	119
China											8	0	10	23	0.5	0.2	
		SPR IAE	O	3.5	1984	-	EK-10	UO ₂ -Mg	Al	10	128	43	45	234	299.5	30	819
Czech. Rep.	Rez	LWR-15															
	Rez	Rez	O	10	1957	-	EK-10	UO ₂ -Mg	Al	10	128	0	19	210	266.8	26.9	
Germany																	
Dresden	RFR	RFR	S	10	1957	1991	EK-10	UO ₂ -Mg	Al	10	128	0	14	144	164.3	18.4	504
Germany																	
Dresden	RAKE	RAKE	S	0	1969	1991	EK-10	UO ₂ -Mg	Al	10	120	0		873	1,048	104.6	3,056
Egypt																	
Inshas	ETR-1	ETR-1	O	2	1961	-	EK-10	UO ₂ -Mg	Al	10	128.7	42	20	50	64.3	6.4	175
Hungary		Training															
Budapest	Reactor	D		0.1	1971	-	EK-10	UO ₂ -Mg	Al	10	128	24	50	28	38	3.6	98
Hungary		Budapest															
Budapest	Reactor	O		10	1959	-	EK-10	UO ₂ -Mg	Al	10	128	0	23	82	105	10.5	287
Hungary																	
Budapest	ZR-6M	O		0	1972	1990		UO ₂ -Mg	Al			0					Decommissioned
DPR Korea																	
Pyongyang	IRT-DPRK	O		8	1965	-	EK-10	UO ₂ -Mg	Al	10	128			44	56.3	5.8	154
Poland																	
Otrock	EWA	S		10	1958	1995	EK-10	UO ₂ -Mg	Al	10	128.6	0	15	184	211	21.1	574

Facility Information				Fuel Assembly Data					Spent Fuel Data							
Country	Facility		Power MW	Critical Date	Shut Date	Fuel Assembly Type	Fuel Meat Type	Clad	Enr. %	Grams U-235 per FA	FA in Core	Average Discharge BU, %	No. Spent Fuel	Total kg U	Total kg U-235	Total Weight of FA
	Name	Status														
Poland																
Otrock	MARYLA	D	0.1	1967	1973	EK-10	UO2-Mg	Al	10	128	0	Decommissioned, 3 fuel tubes, 1 hex, 2 circular				
Romania																
Bucharest	WR-S	S	3	1957	1997	EK-10	UO2-Mg	Al	10	126	0	41	153	195.8	19.6	536
Uzbekistan																
Tashkent	WWR-CM	O	10	1959	-	EK-10	UO2-Mg	Al	10	128.6	0	30	11	14.1	1.4	39
Totals																
2128 2581.3 260.3																
U-Cr-Ni																
Belarus	IRT-M															
Minsk	Minsk	S	4	1962	1988	Rods	U-Cr-Ni	SS	45	173	0		106	40.8	18.3	
Totals																
106 40.8 18.3																
U ₃ Si ₂ -Al																
Chile													0	0	0	0
Santiago	La Reina	O	5	1974	-	MTR	U3Si2-Al	Al	19.8		2					
Totals																
0 0 0																
UO ₂ -Al																
Czech. Rep.		LWR-15														
Rez	Rez	O	10	1957	-	IRT-2M	UO2-Al	Al	36	224	47	40	0	0	0	0
Czech. Rep.		VR-1 Vrabec				IRT-2M	UO2-Al	Al	36	230	7	0	0	0	0	0
Prague	Crit. Fac.	O	0	1990	-	IRT-3M	UO2-Al	Al	36	305	7	0	0	0	0	0
						IRT-3M	UO2-Al	Al	36	355	2	0	0	0	0	0
Germany																
Dresden	RFR	S	10	1957	1991	WWR-MM2	UO2-Al	Al	36	40	0	42	629	69.9	26.2	568
Germany																
Dresden	RAKE	S	0	1969	1991	WWR-M	UO2-Al	Al	36	40	0		77	8.6	3.1	69
Germany																
Germany	ZILFR	O	0	1979	-	WWR-2M	UO2-Al	Al	36	39	94		96	10.3	3.7	86
Hungary		Budapest														
Budapest	Reactor	O	10	1959	-	WWR-M2	UO2-Al	Al	36	44						
DPR Korea						IRT-2M	UO2-Al	Al	36	228			7	4.4	1.6	22
Pyongyang	IRT-DPRK	O	8	1965	-	IRT-2M	UO2-Al	Al	36	198		3-tube (198 g U235), 4-tube (228 g U235)				
Latvia						IRT-3M	UO2-Al	Al	90	270	0	45	13	3.9	3.5	48
Riga	IRT-2M	S	5	1961	1998	IRT-3M	UO2-Al	Al	90	305	0	45	12	4.1	3.7	52
Poland																
Otrock	EWA	S	10	1958	1995	WWR-M2	UO2-Al	Al	36	44.8	0	40	445	55.4	19.9	401
Poland																
Otrock	ANNA	D	0	1963	1997	3-Tube	UO2-Al	Al	21	250	0					Decommissioned

Facility Information					Fuel Assembly Data					Spent Fuel Data						
Country	Facility		Power MW	Critical Date	Shut Date	Fuel Assembly Type	Fuel Meat Type	Clad	Enr. %	Grams U-235 per FA	FA in Core	Average Discharge BU, %	No. Spent Fuel	Total kg U	Total kg U-235	Total Weight of FA
	Name	Status														
Poland																
Otrock	MARIA	O	30	1974	-	Multi-Tubes	UO2-Al	Al	36	550	11	40	1	1.5	0.5	0
Poland																
Otrock	MARYLA	D	0.1	1967	1973	WWR-CM	UO2-Al	Al	36	48.9	0					
Ukraine						WWR-M5	UO2-Al	Al	90	64	0	50	15	1.1	1	14
	Kiev	WWR-M	O	1960	-	WWR-M7	UO2-Al	Al	90	68	0	50	10	0.8	0.7	9
Ukraine																
Sevastopol	IR-100	O	0.2	?	-	EK-10	UO2-Al	Al	10	56	47					
Uzbekistan						IRT-3M	UO2-Al	Al	36	309	12	45	15	12.9	4.6	58
Tashkent	WWR-CM	O	10	1959	-	IRT-3M	UO2-Al	Al	36	352	0					
						IRT-4M	UO2-Al	Al	20	285	2		0	0	0	
						IRT-4M	UO2-Al	Al	20	324	2		0	0	0	
Yugoslavia																
Belgrade	RA	S	6.5	1959	-	TVR-S	UO2-Al	Al	80	7.69	480	30	894	8.8	8.9	
Yugoslavia																
Belgrade	RB	O	0	1958	-	TVR-S	UO2-Al	Al	80	6.67						

UO₂

Czech. Rep.																
Rez	LR-0	O	0	1982	-	Rodded FA	UO2	Zr	2.4.4		63					
Totals																

Polyeth

Germany																
Dresden	AKR	O	0	1978	-	Disc	Polyeth	Poly	19.8	0.93			39	49.5	9.8	57
						Disc	Polyeth	Al	19.8	0.78			1	0	0	0
Totals													40	49.5	9.8	

U-Zr

Kazakhstan																
	EWG 1	O	60	1972	-	Rods	U-Zr	Zr	90							
Totals																

U Metal

Yugoslavia																
Belgrade	RA	S	6.5	1959	-	TVR-S	U Metal	Al	2	7.65	0	37	6,656	2,545	50.9	
Yugoslavia						TVR-S	U Metal	Al	2	7.7						
Belgrade	RB	O	0	1958	-	TVR-S	U Metal	Al	2	7.7						
Totals													6,656	2,545	50.9	

Facility Information			Fuel Assembly Data					Spent Fuel Data								
Country	Facility		Power MW	Critical Date	Shut Date	Fuel Assembly Type	Fuel Meat Type	Clad	Enr. %	Grams U-235 per FA	FA in Core	Average Discharge BU, %	No. Spent Fuel	Total kg U	Total kg U-235	Total Weight of FA
	Name	Status														
Other Reactors																
Latvia											0					
	RKS25	S	0	1966	1993											
Slovakia																
Bratislava	VR-1B	C	0.1	-	-											

Overall Totals 19,230 8,267 1019

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