



# Metal Fuel Fabrication Safety and Hazards

TO NRC-HQ-25-17-T-005, Non-LWR  
LTD2

**June 2019**

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PACIFIC NORTHWEST NATIONAL LABORATORY  
*operated by*  
BATTELLE  
*for the*  
UNITED STATES DEPARTMENT OF ENERGY  
*under Contract DE-AC05-76RL01830*

# **Metal Fuel Fabrication Safety and Hazards**

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the U.S. Department of Energy  
under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory  
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## Executive Summary

The U. S. Nuclear Regulatory Commission (NRC) has developed a vision and strategy<sup>1</sup> that is in line with their need to review and regulate a new generation of non-light water reactors (non-LWRs) and to ensure their readiness to effectively and accurately evaluate these technologies. Several implementation action plans (IAPs)<sup>2, 3</sup> have been developed in order to achieve the goals set forth in the NRC's vision and strategy for non-LWR technology. These IAPs detail activities that will be pursued in the near-, mid-, and long-term timeframes to fully realize the NRC's desire to safely achieve effective and efficient non-LWR mission readiness. These activities include establishing guidance for risk-informed and performance-based development of new non-LWR fuel cycle technologies. One identified gap is the availability of hazard review aids associated with uranium metal and alloy processing operations, as hazards present during metal fuel production can differ greatly from those associated with uranium oxide processes.

To aid in the evaluation of hazards associated with these new technologies, PNNL has been tasked with gathering information that identifies the hazards and any associated safety events related to uranium metal and alloy fuel preparation, processing, and fabrication. These hazards include fire, the use of reactive or toxic chemicals, radiation, and nuclear criticality. PNNL has developed this letter report to present a summary of uranium metal and alloy processing hazards and experience drawn from a literature review.

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<sup>1</sup> "NRC Vision and Strategy: Safely Achieving Effective and Efficient Non-Light Water Reactor Mission Readiness," ADAMS Accession No. ML16356A670

<sup>2</sup> "NRC Non-Light Water Reactor Near-Term Implementation Action Plans," ADAMS Accession No. ML17165A069

<sup>3</sup> "NRC Non-Light Water Reactor Mid-Term and Long-Term Implementation Action Plans," ADAMS Accession No. ML17164A173

## Acronyms and Abbreviations

ADAMS	Agency wide Documents Access and Management System
ADU	Ammonium Diuranate, $(\text{NH}_4)_2\text{U}_2\text{O}_7$
ALARA	As Low As Reasonably Achievable
ANSTO	Australia's Nuclear Science and Technology Organisation
BWXT	BWX Technologies
DOR	Direct Oxide Reduction
DU	Depleted uranium
EBR-II	Experimental Breeder Reactor-II
EFL	Experimental Fuels Laboratory
FCCI	Fuel-Cladding Chemical Interaction
FCF	Fuel Conditioning Facility
FFTF	Fast Flux Test Facility
FMF	Fuel Manufacturing Facility
HEU	High-enriched uranium
HIP	Hot Isostatic Pressing
IAEA	International Atomic Energy Agency
IAP	Implementation Action Plan
INL	Idaho National Laboratory
KAERI	Korea Atomic Energy Research Institute
LANL	Los Alamos National Laboratory
LEU	Low-enriched uranium
LTD	Letter of Technical Direction
LWR	Light Water Reactor
MERMAIDS	Mass, Enrichment, Reflection, Moderation, Absorption, Interaction, Density, and Shape
NFPA	National Fire Protection Agency
NRC	Nuclear Regulatory Commission
NU	Natural uranium
PNNL	Pacific Northwest National Laboratory
PRIDE	Pyroprocess-Integrated inactive Demonstration facility
PUMP-F	Plutonium-Uranium Metal fuel Pin fabrication Facility
PUREX	Plutonium Uranium Redox Extraction
PVD	Physical vapor deposition
RADCON	Radiation Control
REPF	Reference Engineering-scale Pyroprocess Facility
RPL	Radiological Processing Laboratory

RSWF	Radioactive Scrap and Waste Facility
SAR	Safety Analysis Report
SDOR	Saltless Direct Oxide Reduction
SFR	Sodium-cooled Fast Reactor
TBP	Tributyl Phosphate, $(\text{CH}_3\text{CH}_2\text{CH}_2\text{CH}_2\text{O})_3\text{PO}$
TCE	Trichloroethylene, $\text{C}_2\text{HCl}_3$
TIG	Tungsten inert gas
TRU	Transuranic
UCNI	Unclassified Controlled Nuclear Information
UPF	Uranium Processing Facility
VIM	Vacuum induction melting



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# 1.0 Introduction

PNNL has been tasked by the NRC to gather information identifying the hazards and associated safety events related to uranium metal and alloy fuel preparation, processing, and fabrication. These hazards include fire, the use of reactive or toxic chemicals, radiation, and nuclear criticality. This report presents information on process steps and hazards associated with uranium metal and alloy fuel production processes and will be used as input for a uranium metal fuel fabrication safety licensing review aid. The information presented here is mainly from older government operations and may be of limited applicability to specific future commercial nuclear metal fuel fabrication operations. Additionally, this report covers operations discussed in the literature and may not cover all processes in some future license application involving new technology. It should be noted that operational hazards can vary with changes in the processes, operations, or scale of processing.

Definitions of key terms related to the fuel fabrication process can be found in Appendix B.

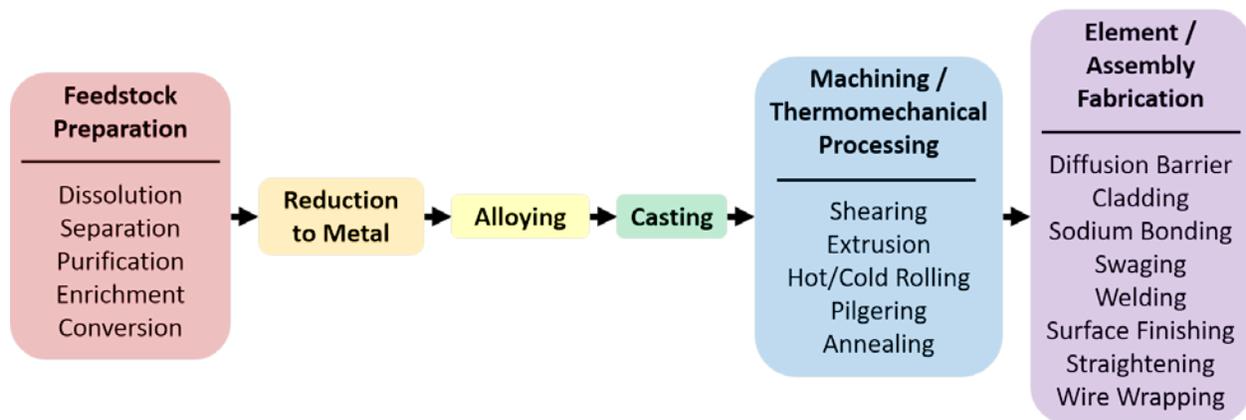
This section presents a brief, high-level overview of a generalized fuel fabrication process—feedstock preparation/scrap recycle, reduction to metal, casting, machining, element/assembly fabrication, and storage—and the associated hazards.

## 1.1 Fuel Fabrication Overview

While uranium metal and alloy fuel fabrication processes are specific to the alloy, reactor type, and even the production facility, they all follow a simplified process flow [1] [2] [3]. In general, the following steps outline the fuel production process:

1. **Feedstock** must be prepared from ore. This includes dissolution, purification, and chemical conversion to the desired chemical state for the next step. Feedstock can also be prepared from used fuel through reprocessing. Enrichment will typically take place between purification and conversion to the final chemical state for reduction but is outside the scope of this effort. (Section 2.0)
2. Feedstock must then be **reduced to metal**. This is traditionally achieved by bomb/metallurgical reduction, but other means can also be employed to convert feedstock to metal. (Section 3.0)
3. The metal is **alloyed** with the desired alloying agent(s) to create a binary, ternary, or other alloy. (Section 4.0)
4. The alloy is **cast** to form a fuel billet. (Section 4.0)
5. The fuel billet is **machined and/or thermomechanically processed** to get it into a desired form. (Section 5.0)
6. The formed fuel billet is **clad and collected into fuel assemblies**. (Section 6.0)

**Figure 1.1** presents a generalized process diagram for metal fuel fabrication. Process flow diagrams that present a highly detailed representation of an example fuel production process can be found in Hubbard et al. [4].



**Figure 1.1.** Generalized process diagram for metal fuel fabrication. Feedstock preparation, machining/thermomechanical processing, and element/assembly fabrication steps list several sub-processes, any or all of which may be included in a fuel fabrication operation.

## 2.0 Feedstock Preparation

Chemical purification and refinement of feedstock, most commonly by ion exchange or solvent extraction, may be performed prior to reduction. This section describes the preparation of feedstock for conversion to metal, including dissolution and any chemical conversions that are necessary to get the feedstock into an appropriate chemical state, and identifies its various hazards. Scrap recovery and recycle is also included here, since the processes are very similar, if not identical, to feedstock preparation.

### 2.1 Process Description

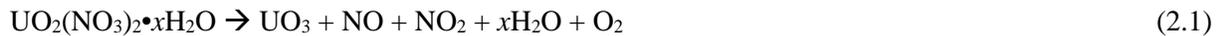
Feedstock preparation for conversion to metal follows the same general flow as that for uranium oxide,  $U_3O_8$ , fuels used throughout the US nuclear reactor complex. The processes for both ore and fuel reprocessing are summarized below.

#### 2.1.1 Feedstock Preparation from Ore

After mining and leaching from ore, uranium is left in the chemical form of uranyl sulfate anion  $[UO_2(SO_4)_3]^{4-}$  when leached with sulfuric acid (the most commonly employed leachate) or uranyl carbonate anion  $[UO_2(CO_3)_3]^{4-}$  when leached with sodium carbonate (primarily used when basic minerals like calcite or dolomite are present in significant quantities). The uranium complex can then be either loaded onto an ion exchange column or purified via solvent extraction. After this step, uranium is generally precipitated as ammonium diuranate (ADU),  $(NH_4)_2U_2O_7$ , commonly referred to as yellowcake.

Yellowcake can be further purified by dissolution in nitric acid and undergoing solvent extraction using tributyl phosphate (TBP) mixed with kerosene. Solvent extraction can be performed using centrifugal contactors, pulse columns [5], or mixer-settlers [6], all of which work on the same general principal of intimately mixing the aqueous and organic phases then allowing them to settle out and separate.

Uranium is then stripped from the TBP in the form of uranyl nitrate,  $UO_2(NO_3)_2$ . Uranyl nitrate can be denitrated and calcined to  $UO_3$  (Eq. 2.1) and then further reduced to  $UO_2$  using hydrogen gas (Eq. 2.2) [6].



$UO_2$  can be converted to  $UF_4$  via either a wet (Eq. 2.3) or dry (Eq. 2.4) fluorination process using either aqueous or anhydrous HF [7] [6]. In the wet fluorination process, the product must be further dried to ensure most of the water is removed prior to conversion to metal. In the dry fluorination process, the reaction is generally achieved in a fluidized bed-type reactor.



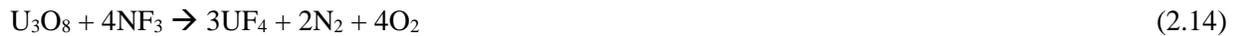
Alternative methods for formation of  $UF_4$  include

- Hydrolysis of  $UF_6$  to  $UO_2F_2$ , then reduction to  $UF_4$  using a reducing agent [8]





- Use of  $\text{NF}_3$  as the fluorinating agent instead of  $\text{HF}$  or  $\text{F}_2$ , with postulated reactions including the following [9]



Typically conversion of  $\text{UF}_4$  to  $\text{UF}_6$  using fluorine gas and enrichment will then occur; however, the enrichment process is beyond the scope of this work.  $\text{UF}_6$  will then be converted back to  $\text{UF}_4$ .

Other final chemical forms, such as uranium bromide, chloride, or iodide, may be required depending on the process chosen for conversion to metal (see Section 3.1). However, these feed materials have unfavorable properties (hygroscopic, expensive) and have not been well-studied [6].

## 2.1.2 Feedstock Preparation from Used Fuel (Fuel Reprocessing)

After removal from a reactor, used fuel can be reprocessed for reuse. Fuel reprocessing can be described two major steps: disassembly of used fuel elements and purification. Specific steps for reprocessing of sodium-cooled fast reactor fuel are summarized below.

Because sodium is highly pyrophoric, used fuel subassemblies from a sodium-cooled fast reactor (SFR) must first be processed in a vessel under an inert atmosphere until the sodium is removed. Sodium removal can be achieved by oxidation with dilute oxygen in nitrogen (limiting the reaction rate), followed by water vapor in nitrogen. The oxidized sodium is then removed by flowing water through the vessel (which will also serve to oxidize any unreacted sodium) [10].

The fuel elements must then be removed mechanically from the subassemblies, generally by shearing, and decanned/declad [10]. Decladding can be done either mechanically [10] or chemically [11] [12]. In mechanical decladding, the two ends of the fuel element are sheared off; the rest of the cladding is then removed by either cutting the cladding on opposite sides to remove the cladding in two pieces or spiral cutting [10]. Chemical decladding method depends whether the cladding is stainless steel or zirconium-based. Dissolution of stainless steel is accomplished via hot 2M hydrochloric acid ( $\text{HCl}$ )-5M nitric acid ( $\text{HNO}_3$ ) (the Darex Process) [12]. Dissolution of zirconium-based cladding is accomplished either via dry hydrogen chloride at 400-600°C (the Zircex Process) [12] or ammonium fluoride ( $\text{NH}_4\text{F}$ ), either alone or mixed with ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) (the Zirflex Process) [11].

The fuel must then be purified to removed fission products from the used fuel, either by melt refining [10], as described in Section 4.1.3, or by dissolution (in hot nitric acid [12], HCl, HCl and hydrogen peroxide, or anodically in ammonium carbonate) and solvent extraction, as described in Section 2.1.1 above.

## 2.2 Pyrophoricity Hazard

Uranium oxide is considered pyrophoric [13]; fires attributed to uranium oxide powders have been previously observed in fuel cycle facilities [14].

Hydrogen gas ( $H_2$ ), used to reduce  $UO_3$  to  $UO_2$ , is highly flammable. Additionally, while oxygen ( $O_2$ , produced as an off-gas during several chemical conversions that are part of feedstock preparation) is not flammable, it can oxidize other materials and cause them to ignite at a lower temperature.

Reprocessing used fuel presents added pyrophoricity hazards. First, used fuel experiences self-heating; EBR-II fuel that underwent 2 a/o burnup in 135 days and was then cooled for 15 days will produce ~200 W for every kg of fuel (assuming that all beta and gamma radiation is absorbed) [10]. Used fuel from SFRs requires special considerations as well, as sodium will spontaneously explode when exposed to water due to formation of hydrogen gas.

## 2.3 Chemical Hazards

The predominant hazard associated with feedstock preparation is chemical due to the many chemical reactions and manipulations necessary to purify the material and obtain a form suitable for conversion to metal.

Uranium fumes and oxide powder are highly toxic, and heavy metal poisoning from inhalation is possible [15] without appropriate controls.

While all acids used throughout the process pose a hazard, one of the most hazardous is HF in both the gaseous and aqueous forms. Gaseous HF poses an immediate, acute hazard to the lungs and corneas if personnel are exposed. Aqueous HF is a contact hazard; it causes deep but painless burns upon contact that can lead to tissue death and also affects calcium metabolism through the formation of calcium fluoride (CaF) and can lead to cardiac arrest and death.

Fluorine ( $F_2$ ) is also highly hazardous; it is an identified poison, oxidizer, and inhalation hazard, causing irritation to the eyes and respiratory system upon exposure. Severe liver and kidney damage can also occur with exposures above concentrations of 25 ppm.

Beyond the immediate hazard to personnel, HF and  $F_2$  are highly corrosive to most metals [6]. Any corrosion to pipes may eventually result in a release.

## 2.4 Radiation Hazard

The biggest radiation hazard of feedstock preparation from ore is exposure to radium and its daughter products. Ores with high radium content may require shielded operation until the feedstock is purified.

Fuel reprocessing presents its own radiation hazards. Used fuel is highly radioactive due to fission products—used fuel from EBR-II requires approximately 11 inches of lead shielding to reduce the radiation level to less than 0.1 mR/hour [10]. Sodium-24 is produced in reactors by neutron bombardment of sodium-

23 and has a half-life of 15 hours [10]; sodium contamination must be monitored in all associated equipment. Uranium-236 must also be considered in used fuel.  $^{236}\text{U}$  is produced when  $^{235}\text{U}$  absorbs a neutron and does not fission (~18% of reactions), is very challenging to remove, and contributes significantly to the radioactivity of used fuel.

Uranium, especially depleted (DU) [15], natural (NU) [16], and low-enriched uranium (LEU), poses a very minor radiation hazard as it is only weakly radioactive. However, DU does emit fairly intense  $\beta$  radiation; inhalation of particulates can result in deposition of  $\beta$ -emitters in the lungs [17]. Employing ALARA (“as low as reasonably achievable”) principles of increased distance and decreased exposure time reduces the radiation hazard presented by the fuel material.

## 2.5 Nuclear Criticality Hazard

Criticality is influenced by several parameters, summarized by the acronym MERMAIDS (Mass, Enrichment, Reflection, Moderation, Absorption, Interaction, Density, and Shape). Because criticality is highly dependent on multiple factors, it is difficult to make an overarching assessment of criticality hazards without knowledge of specific working conditions. Facilities should be evaluated by a criticality safety analyst and criticality safety engineer to ensure safe practices.

Criticality hazard is negligible when using natural uranium [16] without a special moderator (e.g., heavy water). The main criticality risks occur after enrichment and are the unexpected precipitation or settling of uranium compounds and the use of vessels that provide a favorable configuration for criticality.

## 3.0 Reduction to Metal

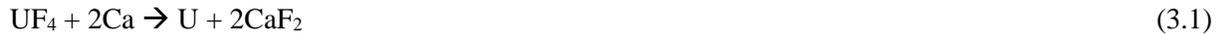
Bomb or metallothermic reduction (generally magnesiothermic or calciothermic) is generally and traditionally used to convert uranium green salt to metal, but reduction to metal may also be achieved via alternative methods. This section describes the various processes that can be used to reduce feedstock/recycled scrap to metal and identifies their hazards.

### 3.1 Process Description

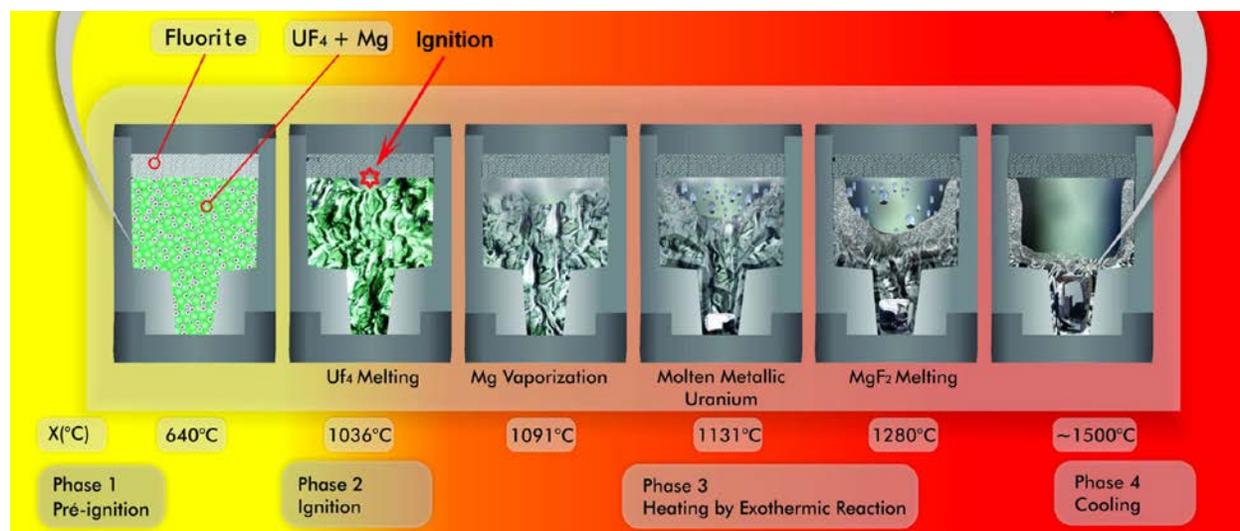
Methods for reduction to metal can largely be classified into one of four general categories: (1) reduction of uranium halides (fluorides, chlorides, bromides, or iodides) by metals (Ca, Mg, Na), (2) reduction of uranium oxides by metals (Al, Ca, Mg) or carbon, (3) electrolytic reduction of uranium oxide, and (4) disproportionation or thermal decomposition of uranium halides [6]. Of these, reduction of halides by metals is the most popular method as it produces a large metallic mass, while the other methods produce uranium shot or powder [6]. A description of each category follows.

#### 3.1.1 Reduction of Uranium Halides by Metals

Of the various methods for reduction of halides by metals, calciothermic reduction of  $UF_4$  is commonly employed due to its high exothermic heat [18]. However, magnesiothermic reduction of  $UF_4$  has several advantages over calciothermic reduction, including avoiding the use of toxic and pyrophoric calcium and use of a cheaper reducing agent [18]. A charge is prepared by mixing together  $UF_4$  and the reactant homogeneously in a crucible made of refractory material (generally magnesium oxide or graphite); the crucible is then capped, put into a reaction vessel (generally steel), and surrounded by additional refractory (magnesium oxide) sand. The calciothermic and magnesiothermic reactions proceed via similar reactions:



Both reactions require pre-heating of the reactants (with magnesium needing higher heat than calcium) and a stoichiometric excess of the reducing agent to ensure the reaction runs to completion. Pre-heating can be accomplished via an induction coil, pit furnace [18], or more uniquely, a laser pulse [19]. The charge ignites and runs; after cooling, the uranium metal can be separated from the  $CaF_2$  or  $MgF_2$  slag. **Figure 3.1** presents an illustration of the sequence of events for magnesiothermic reduction of  $UF_4$ , taken from Durazzo et al. [18].

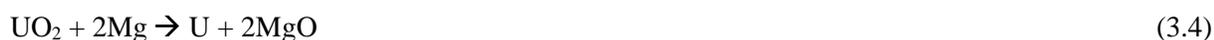


**Figure 3.1.** Illustration of magnesiothermic reduction of  $UF_4$  to U metal (from Durazzo et al. [18]).

Uranium chlorides, bromides, and iodides can also be bomb reduced with metals, but these feed materials have unfavorable properties (hygroscopic, expensive) and have not been well-studied [6].

### 3.1.2 Reduction of Uranium Oxides by Metals

Direct Oxide Reduction (DOR), also known as Saltless Direct Oxide Reduction (SDOR) [5], is a method to reduce uranium oxides (mainly  $UO_2$ , but also  $U_3O_8$  or  $UO_3$ ) by various metals (Ca, Mg, Al) and also by carbon [6]. Use of an iodine booster in calcium reduction of  $UO_3$  can produce a metallic mass (~30% of uranium) with the rest powder [6]. DOR requires very high temperatures to initiate reduction, especially when reducing with carbon [6], but does not require fluorination of the feedstock or use of a pressure vessel as in bomb reduction [5]. Otherwise, the reactions are very similar:

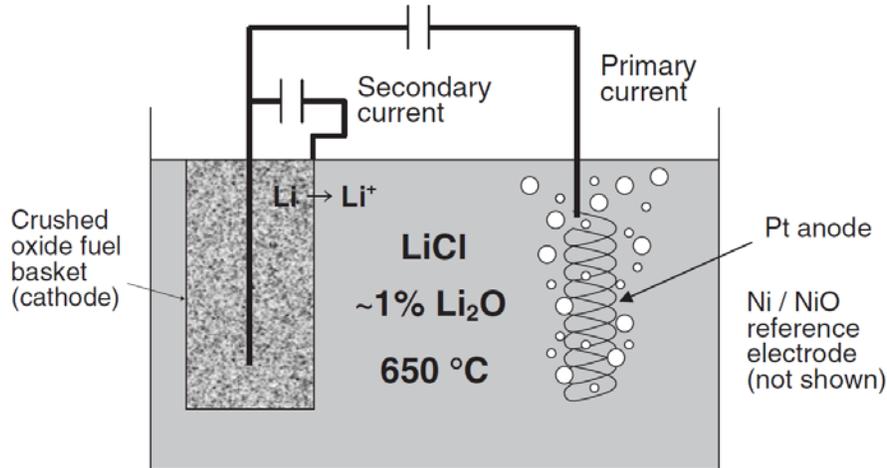


### 3.1.3 Electrolytic Reduction

In electrolytic reduction, oxide fuel is loaded into a cathode basket immersed in molten  $LiCl-Li_2O$  with a platinum wire acting as anode. Upon application of an electric current, the reaction



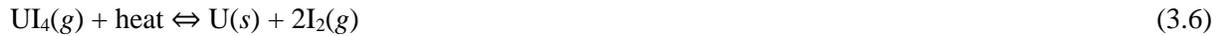
occurs, with the uranium metal retained at the cathode and the  $Li_2O$  released into the molten salt [20]. **Figure 3.2** presents an illustration of the electrolytic cell used for reduction of  $UO_2$  to U metal, taken from Herrmann et al. [20].



**Figure 3.2.** Illustration of electrolytic cell for reduction of  $\text{UO}_2$  to U metal (from Herrmann et al. [20]).

### 3.1.4 Disproportionation or Thermal Decomposition of Uranium Halides

Disproportionation or thermal decomposition is a temperature-dependent reaction that can result in very pure uranium metal according to the following (reversible) equations [6]:



In these reactions, a heated vessel with a hot filament is filled with a uranium halide. The filament provides the heat to make the reactions proceed to the right, while in the cooler parts of the vessel the reaction proceeds to the left [6]. These reactions, especially (3.7) and (3.8), require very high temperatures and have not been historically used on a commercial basis.

## 3.2 Pyrophoricity Hazard

The main pyrophoricity hazard arises from uranium metal as it poses a significant fire hazard. The Frank-Kamenetskii parameter  $\delta$  can be used to determine the conditions for ignition:  $\delta \geq 1$  suggests ignition, with  $\delta$  defined as

$$\delta = \frac{V \cdot \Delta H \cdot C \cdot T_r}{h \cdot A \cdot T_\infty} e^{-T_r+1}, \quad (3.9)$$

where  $V$  is the volume,  $\Delta H$  is the effective heat of reaction per mass,  $C$  is a pre-exponential factor in the Arrhenius form of the material consumption rate,  $T_r$  is the ratio of the activation temperature  $T_E$  to the ambient temperature of the piece  $T_\infty$ ,  $h$  is the overall heat transfer coefficient, and  $A$  is the surface area [21]. A smaller piece of material (and therefore smaller  $A$  and  $V$ ) requires a higher ambient temperature for ignition. Also, use of coolant can increase  $h$  over the minimum value required to avoid conditions for ignition. Other variables are more or less constant for a given material system or process.

Uranium metal may also be entrapped within the slag produced during reduction [18], requiring that slag be treated and stored with care.

Calcium [18] and magnesium [13] are also highly pyrophoric metals.

### **3.3 Chemical Hazards**

Both calcium and uranium are toxic chemicals [18]. Uranium in particular poses a hazard from inhalation of the powdery oxide form. Heavy metal poisoning from inhalation is possible [15] without appropriate controls.

### **3.4 Radiation Hazard**

Uranium, especially DU [15], NU [16], and LEU, poses a very minor radiation hazard as it is only weakly radioactive. However, DU does emit fairly intense  $\beta$  radiation; inhalation of particulates can result in deposition of  $\beta$ -emitters in the lungs [17]. Employing ALARA principles of increased distance and decreased exposure time reduces the radiation hazard presented by the fuel material.

Care must also be taken to prevent dispersion and contamination;  $UF_4$  and uranium oxide powders can be challenging to contain and can create inhalation hazards.

### **3.5 Nuclear Criticality Hazard**

Very large reductions of natural uranium can be performed (100 kg [18] or more) without criticality concerns. Low-enriched uranium (LEU), however, requires the reduction of smaller charges to prevent criticality—nominally 2.2 kg U or less [18]. Higher enrichments necessitate the reduction of even smaller charges.

## 4.0 Alloying and Casting

Uranium metal must often be alloyed to achieve properties for improved performance as reactor fuel [6], including corrosion resistance, hardness, and phase stability. Various binary (e.g., U-Mo [4], U-Zr [22]) and ternary (e.g., U-Pu-Zr [22]) alloys have been explored to meet these needs. Fission, or fission products from used nuclear fuel nominally composed of 49.2wt.% Mo, 39.2wt.% Ru, 5.6wt.% Rh, 3.8wt.% Pd, 2wt.% Zr, and 0.2% Nb, has also been explored as an alloying agent to produce U-5%Fs [22]. In addition to alloying, casting allows the ability to produce uranium in a desired shape (e.g., plate or rod) [5]. This section describes various alloying and casting processes employed to form fuel elements and identifies their associated hazards. Hazards may vary depending upon the casting method; method-specific hazards are noted in Sections 4.2 through 4.6.

### 4.1 Process Description

Several methods have been used to alloy and cast uranium fuel:

- Co-reduction,
- Arc melting,
- Vacuum induction melting (VIM) and injection casting, or casting via pressure injection into a mold,
- VIM and gravity-fed injection into a mold, and
- Microwave casting, or use of a microwave furnace to melt and cast.

The following sections present a brief description of each process, as well as a summary of other casting methods that have been deemed less desirable for uranium casting.

#### 4.1.1 Co-reduction

The bomb reduction technique described in Section 3.1.1 can also be used to co-reduce a compound of the alloying agent (generally a fluoride) with  $UF_4$  [6]. The alloying agent compound is mixed to create a homogeneous mixture with the  $UF_4$  and reducing agent which is then reduced, resulting in a metallic alloy that can either be recast using one of the below methods or be used as-is for fabrication of fuel elements.

#### 4.1.2 Arc Melting

Arc melting can be carried out either with a tungsten (non-consumable) electrode or with a consumable electrode containing the desired alloying agent and results in a high-purity, homogeneous alloy due to the high temperatures achievable. In either case, arc melting is performed within an airtight vessel that is evacuated and then inerted to minimize contamination. A water-cooled crucible (generally copper) acts as the anode, while the electrode serves as the cathode. A DC current is passed between the electrode and crucible, melting the loaded alloy charge (in the case of non-consumable electrode) or both the uranium in the crucible and the consumable electrode. The electrode can also be a composite of both the uranium and alloying agent as a “bundle” [6] or sandwich [23], in which case both metal electrodes are consumed during melting. Arc melting is generally only employed to form a “master alloy” which is then cast into the desired

shape using one of the following methods [22]. Vacuum arc remelting (VAR) may also be necessary when there is a large difference in melting point for the various fuel components [24].

### **4.1.3 Vacuum Induction Melting**

VIM is historically and currently the most widely used method for fuel fabrication [15] [6] [25] [26] [27]. Heating is achieved by an induction, or work, coil through which an alternating current is passed; a magnetic field is formed, inducing a current and generating heat by electrical resistance in the crucible or material to be melted [26]. Crucible loading is crucial to reduce the loss of volatile elements: the element with the lowest melting temperature (e.g., Pu) should be loaded into the bottom, with U on top so that as it melts it encapsulates the other alloying agent(s) [25]. Charges are commonly melted in yttria-coated graphite crucibles [25] [28]. Homogenization of the melt is achieved by inductive flux field stirring [15] and convection [22].

In injection casting, molds are lowered into the crucible below the melt line, allowed to heat, then the furnace is rapidly pressurized using argon gas [25] to force the melt into the molds via the pressure differential between the inside and the outside of the mold [28]. Refractory-coated quartz [25] [29] [28] or Vycor [22] molds are traditionally employed; as these are not reusable, this can create a significant waste stream. Originally injection casting was achieved via two separate melts—the charge was first alloyed and poured into ingots, then the ingot was melted and injection casted—but the addition of a stirring paddle allowed the casting to be completed in a single step [22]. Preheating the molds can also improve cast quality [22].

Gravity-fed casting can be achieved via either bottom pour [25] [4] [26] or tilt-pour methods. In bottom pour casting, the crucible is held above the mold in such a manner that neither the crucible nor the mold must be moved to pour the molten metal [26]. After the charge has been melted and held at temperature for the prescribed time, a knockout plug is punched out and molten metal flows into the mold [4]. Tilt-pour casting is similarly achieved, except instead of the removal of a knockout plug the crucible is tilted (up to 90°) to pour into a mold. Tilt-pour tends to result in a more homogeneous product as lighter elements tend to float to the top of the melt and pouring provides mixing [26]. Yttria-coated graphite molds are generally used for gravity-fed casting.

Melt refining is a special application of VIM for fuel reprocessing, in which bare used fuel elements are melted, held at temperature for an extended period of time (one to three hours) to allow volatile fission products to be removed as gas, and then cast into a mold, leaving reactive fission products as a casting skull in the crucible [10]. The purified, cast material can then either be used as-cast, or recast as necessary or desired.

### **4.1.4 Microwave Casting**

Microwave casting is being explored as an alternative to more traditional induction melting and casting processes. Instead of induction heating, microwave casting uses microwave energy coupled to a ceramic crucible to melt a charge and cast into the desired form [5]. The main advantage over VIM casting is decreased carbide and oxide formation [5]. However, this method is not as well-developed as VIM casting. Additionally, while induction provides some electromagnetic stirring to help homogenize alloys, microwave casting is a more stagnant process and may require advanced alloying through arc melting or addition of a stirring mechanism.

### 4.1.5 Others

Some other casting methods include centrifugal casting, continuous casting, and cold crucible casting [25]. Centrifugal casting is achieved by a vacuum induction furnace mounted over a centrifuge [1] and was determined to be undesirable due to its complex equipment and small casting diameter [22]. Continuous casting was achieved at the Korea Atomic Energy Research Institute (KAERI) using a feeder that continuously fed material into a tilt-pour type melt crucible, which poured into several molds on a turntable [30].

## 4.2 Pyrophoricity Hazard

Choice of coil for VIM is of great importance to reduce resistive heating and to serve as a heat sink; solid, oxygen-free, high-conductivity copper is preferable [25]. The use of a solid copper bar instead of hollow tubing reduces resistive heating [22].

As previously stated, uranium metal poses a significant pyrophoricity hazard (see Section 3.2). Uranium metal can be retained in the casting skull (metal that is incompletely drained and freezes inside the crucible [4]) and therefore must be treated accordingly, either recycled via the process in Section 7.0 or stored in a safe manner.

Alloyed uranium is less reactive than pure uranium metal; it has been demonstrated that U-10wt% Mo oxidizes more slowly than pure U [31].

## 4.3 Chemical Hazards

Calcium can be used for co-reduction. Both calcium and uranium are toxic chemicals [18]. Uranium in particular poses a hazard from inhalation of the powdery oxide form.

Magnesium can also potentially be used for co-reduction and is pyrophoric, as noted in Section 3.2.

## 4.4 Radiation Hazard

Uranium, especially DU [15], NU [16], and LEU, poses a very minor radiation hazard as it is only weakly radioactive. However, DU does emit fairly intense  $\beta$  radiation; inhalation of particulates can result in deposition of  $\beta$ -emitters in the lungs [17]. Employing ALARA principles of increased distance and decreased exposure time reduces the radiation hazard presented by the fuel material.

Any uranium metal fuel alloyed with transuranics (TRU) requires fabrication in a hot cell facility [2] or appropriate glovebox [32] where remote handling can be achieved due to the high radiotoxicity and radioactivity of TRU elements.

Dispersion of radiation during the casting process, especially more volatile elements [29] like americium [25] or plutonium, can be of concern and may compel the use of ventilation or a glovebox during casting [33] [22].

Plutonium presents an increased contamination risk over uranium. Contamination leaks have been discovered during previous casting activities involving Pu-containing alloys, including during bagout activities from a glovebox and from leaks in piping that crossed the glovebox containment [22].

Uranium daughter products are highly radioactive; much of this material will be retained at the top of the casting (the so-called “hot top”) and is treated as waste [27].

Melt refining creates both a highly radioactive gas of volatile fission products and a highly radioactive casting skull of reactive fission products. This necessitates the use of a hot cell facility for both protection from radiation and prevention of contamination [10].

## **4.5 Nuclear Criticality Hazard**

Water cooling of induction coils can present a criticality hazard [25] [22].

Criticality is the hazard with the most variability between casting methods; this is controlled mainly by limiting the mass of a charge. Criticality safety studies conducted by Stephen and Reddy [34] determined that the maximum safe fissile mass in an induction melting furnace is 10.5 kg. However, criticality also depends on geometry, moderation, reflection, neutron absorption, and concentration and therefore fissile mass limits must be evaluated for each melting setup. The possibility of “mis-pours” must also be considered, particularly for bottom-pouring configurations and continuous casting. Crucible cracking or metal freezing may cause metal to escape the crucible-mold setup and must have a criticality-safe configuration to flow into. Metal freezing may similarly cause metal to stay in the crucible.

## **4.6 Electrical Hazard**

Induction coils can present a significant electrical hazard. Electrical arcing can happen due to coil spacing or problems with the coil insulator. Fuel dust must also be cleaned from the furnace to avoid corona arcing. Not only is this arcing detrimental to the melt process, it can damage the power supply or cause a complete failure. The corona effect is a function of coil voltage, coil surface temperature, chamber pressure (during injection casting), and the dielectric properties of surrounding materials [22], emphasizing the importance of coil material.

## 5.0 Machining and Thermomechanical Processing

This section describes the various machining and thermomechanical processes, including shearing, extrusion, rolling, hot working, and cold working, that are performed on bare uranium metal and alloy elements. All processes related to adding cladding to the fuel elements and those performed on clad fuel elements are discussed in the next section (Section 6.0).

### 5.1 Process Description

Fuel can be processed by several machining and thermomechanical methods prior to cladding and further manipulation. First, the hot top must be removed, either by a cut-off saw [35], commercial shears [22], a hammer and chisel [22], or other method. This removes impurities from the fuel piece. The removed hot top is then treated as scrap.

Fuel can be heated in a vacuum annealing furnace to ensure homogenization of the alloying agents [4] [23].

The fuel billets are then machined via a variety of methods. The billets can be machined on a tracer lathe with a pair of form cutters [35]. The fuel can also be hot rolled: the fuel is canned in steel hot rolling cans and welded using tungsten inert gas (TIG) welding to seal the can, heated, and run through the rolling mill multiple times for decreasing gap settings to achieve the desired thickness [4]. After hot rolling the fuel is annealed, sheared to open the can and remove the fuel, then cold rolled in either gap (set distance between rollers) or force (rollers touching with the amount of force between the rolls controlled) mode to achieve final thickness [4]. In general, hot rolling is used for large reductions, while cold rolling is used for finishing reductions. The fuel is then vacuum annealed to relieve stresses from cold work and sheared to the desired size [4].

Uranium and its alloys can be extruded to produce rods using molten glass as a lubricant and protective coating [23]. Extrusions are accomplished with either horizontal or vertical hydraulic presses [23].

Immersion of the fuel in a salt bath prior to hot working provides for more uniform heating and oxidation protection during heating [4]. The salt bath may be composed of chlorides and/or carbonates (lithium and barium carbonates, lithium and potassium carbonates) [15]. Alternatively, infrared heating [5] or a muffle furnace can be used to preheat and anneal. If canning is employed, temperature must be limited to prevent the formation of low melt temperature eutectics (e.g., U-Fe).

Finally, the fuel must be degreased and decontaminated. Degreasing can be done with trichloroethylene (TCE) [35], Triton DF-16 [4], or a similar degreasing agent. Decontamination can then be completed by wiping the fuel with an alcohol-soaked lint-free cloth [4], or by etching in sodium hydroxide (NaOH) and then a nitric acid-hydrofluoric acid mixture [35].

### 5.2 Pyrophoricity Hazard

Pyrophoricity poses an even stronger hazard during fuel cutting, machining, and shearing (see Section 3.2). Uranium fines and chips will burn and can generate a violent exothermic reaction with water [17]. Work that goes into forming a chip or fines shows up as heat in the work piece; there is also a friction interface between the chip and tool that generates heat. Faster machining generates more heat [17].

The hot top contains uranium metal or alloy and therefore must be either recycled via the process in Section 2.0 or stored in a safe manner.

Dissolution of NaOH in either water or acid is a highly exothermic reaction, producing significant heat.

Both TCE [36] and Triton DF-16 [37] have a National Fire Protection Agency (NFPA) 704 flammability rating of 1, meaning that they require considerable preheating before ignition/combustion occurs.

### **5.3 Chemical Hazards**

Uranium can be a chemically active material, which may require special fluids to use as lubricant or coolant during machining and cutting operations [17].

TCE, used for degreasing [35], can depress the central nervous system and was previously used as an anesthetic. It may also cause cancer and is suspected of causing genetic defects [36]. Therefore, worker exposure and inhalation is highly undesirable.

Triton DF-16 can cause moderate eye irritation and severe corneal injury upon eye contact and moderate skin irritation upon skin contact. However, it poses no inhalation hazard [37].

NaOH, used to etch and decontaminate, is a caustic chemical and can cause chemical burns. Nitric acid and HF are also used to etch (see Section 2.3 for details on HF hazards).

### **5.4 Radiation Hazard**

Uranium, especially DU [15], NU [16], and LEU, poses a very minor radiation hazard as it is only weakly radioactive. However, DU does emit fairly intense  $\beta$  radiation; inhalation of particulates can result in deposition of  $\beta$ -emitters in the lungs [17]. Employing ALARA principles of increased distance and decreased exposure time reduces the radiation hazard presented by the fuel material.

Any uranium metal fuel alloyed with transuranics (TRU) requires fabrication in a hot cell facility [2] or appropriate glovebox [32] where remote handling can be achieved due to the high radiotoxicity and radioactivity of TRU elements.

Machining processes may generate radioactive dust [17], which can then be dispersed and result in significant room contamination [5]. This may compel the use of ventilation or gloveboxes for machining and thermomechanical processing.

### **5.5 Nuclear Criticality Hazard**

Criticality is only of concern if adequate fuel spacing is not maintained. Care must also be taken to avoid the buildup of uranium fines/chips to prevent formation of a critical mass. Because criticality is highly dependent on multiple factors, it is difficult to make an overarching assessment of criticality hazards without knowledge of specific working conditions. Facilities should be evaluated by a criticality safety analyst and criticality safety engineer to ensure safe practices.

## 6.0 Element/Assembly Fabrication

Processes involving cladding application, finishing steps, and fabrication of full fuel assemblies are described in this section.

### 6.1 Process Description

Often, a diffusion barrier (usually zirconium) is necessary between the fuel and cladding to prevent fuel-cladding chemical interaction (FCCI),<sup>1</sup> which can form undesirable compounds that affect fuel performance [38]. Methods for diffusion barrier application include co-rolling, electroplating, physical vapor deposition (PVD), or plasma spraying [4]. While these methods would work for annular shaped fuel, a zirconium sheath may alternatively be applied by casting with a zirconium tube in the mold or placing a thin tube into the cladding before inserting the fuel slug [39].

Since element fabrication and fuel assembly construction are highly dependent upon the final fuel form (plate, rod, tube, etc.) and the reactor for which the fuel will be used, a few different procedures are described. Those described here are not all-inclusive but rather represent examples of process flows.

Some other machining processes that may take place during element and assembly fabrication include roll/friction bonding, swaging, surface finishing, and plate straightening.

#### 6.1.1 Rod-Type Fuel

##### 6.1.1.1 Rod-Type Fuel Elements for Sodium-Cooled Fast Reactors

Production of rod-type fuel elements for sodium-cooled fast reactors is described in Batte [33]. Briefly:

Jackets are produced by welding the lower end plug to stainless steel tubing. A stainless steel wire is then welded to the lower end and wrapped in a machine at a specified pitch to the top of the tubing, where the wire is welded to the top of the tube.

The jackets are then loaded with sodium and the fuel slugs inserted. The loaded jackets are then put into a settling furnace, brought to temperature, and the slugs settled into the molten sodium to bond the slug to the jacket. The fuel element can be loaded with a tag gas (to identify when a fuel element is breached and which element it is) or an inert gas. Then, the end plug is inserted and welded. All jacket loading operations are conducted within an inerted glovebox.

Sub-assemblies are manufactured by bundling the fuel elements and inserting them in stainless steel hexagonal tubing. The hexagonal tubing is enclosed with upper and lower plug assemblies.

##### 6.1.1.2 Plutonium Rod-Type Fuel

Production of plutonium fuel rods is described in Bailey et al. and summarized in **Figure 6.1** [35]. Briefly:

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<sup>1</sup> Diffusion barriers are only required when cladding is not Zr-based, as is the case with fast reactor fuels (stainless steel cladding) or research reactor fuels (Al cladding).

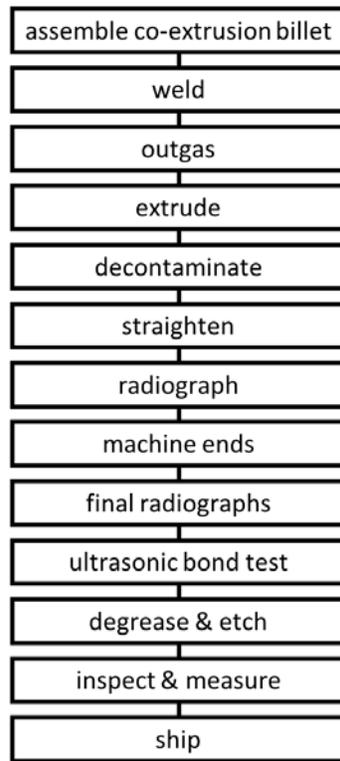
The trailing-end plug is fusion welded to the fuel billet and an aluminum tube is welded to the trailing-end plug protrusion. This unit is then outgassed at an elevated temperature. The tube is sealed by pressure welding using modified bolt cutters.

Plutonium alloy cores clad with aluminum alloy were co-extruded. The billet is placed in the extrusion press furnace at elevated temperature and lubricated with a graphite-oil dispersion. The extrusion press used had a streamline flow-type die with a 90° entrance angle. Use of a guide tube on the extrusion press improved the straightness of the resulting tube. An overall reduction of 5.9 to 1 in the core and 11 to 1 in the cladding was achieved (with the variation due to a hardness difference). The fuel rod was then trimmed to facilitate handling and fabrication and etched in sodium hydroxide and then nitric-hydrofluoric acid.

Fuel rods were processed through a rod straightener; the number of passes required was decreased by using the guide tube during extrusion. Kerosene was used as a lubricant.

The fuel rods were cut to length and plug and socket-type end fittings were machined onto the rods.

Finally, the fuel rods were degreased and etched using sodium hydroxide and nitric-chromic-hydrofluoric acid.



**Figure 6.1.** Process flow diagram for fabrication of co-extruded Al-Pu fuel elements [35]

### 6.1.2 Plate-Type Fuel

Production of fuel plates is described in Hubbard et al. [4]. Briefly:

Cladding is applied via hot isostatic pressing (HIP). Fuel (with a Zr diffusion barrier applied) is sandwiched by aluminum cladding plates and placed in several layers separated by a strongback then placed in a HIP can. The can is tack welded, completely sealed by electron beam or TIG welding, and leak checked using

helium. This package is then pressed to join the aluminum cladding to the fuel. The HIP can is opened by shearing to extract the clad fuel packages.

Excess cladding around the edges of the fuel is sheared off. The cladding is thinned by chemical milling until the desired thickness is achieved. The plates are then formed as necessary according to the requirements for each reactor.

Plates are stacked into welded fuel assembly, end caps are placed and welded in place, an anti-corrosion coating is applied as necessary, and the assemblies are packaged and shipped.

## **6.2 Pyrophoricity Hazard**

Pyrophoricity of uranium is not as much of a concern for clad fuel elements, as the uranium is not readily exposed to air or moisture.

Aluminum and zirconium can pose a pyrophoricity hazard when finely divided [13] (i.e., fines, powder).

Sodium, used for fuel-cladding bonding in fuel elements for sodium-cooled fast reactors [33], will spontaneously explode when exposed to water due to formation of hydrogen gas.

## **6.3 Chemical Hazards**

Sodium, used for fuel-cladding bonding in fuel elements for sodium-cooled fast reactors [33], can cause chemical burns.

NaOH, used to etch and decontaminate, is a caustic chemical can cause chemical burns. Nitric acid and HF are also used to etch (see Section 2.3 for details on HF hazards).

## **6.4 Radiation Hazard**

Application of end fittings for the plutonium fuel rods was performed in an open-front hood; no contamination was observed [35].

## **6.5 Nuclear Criticality Hazard**

Criticality is only of concern if adequate fuel element or assembly spacing is not maintained. Because criticality is highly dependent on multiple factors, it is difficult to make an overarching assessment of criticality hazards without knowledge of specific working conditions. Facilities should be evaluated by a criticality safety analyst and criticality safety engineer to ensure safe practices.



## 7.0 Scrap Recycle

Scrap recycle follows the same general procedure as feedstock preparation from ore (described in Section 2.0), except that the dissolution procedure can vary. Sources of uranium-bearing scrap include  $\text{CaF}_2$  resulting from calciothermic reduction of  $\text{UF}_4$  [40] (or slag resulting from other reduction methods), filter cakes from the purification process [40], floor washings [41], oxides, and metal chips/turnings/etc. from casting, machining, and thermomechanical processing [5].

Oxide scrap (from casting skulls or other oxidized scrap from cutting/machining fines and chips) can be leached in the same way as ore, but metal dissolution requires a few more steps. Cladding must be removed if present (using the Zirflex [11] or Zircex [12] processes for zircaloy or other zirconium-based cladding). The uranium can then either be ground and calcined to an oxide form, dissolved in hot nitric acid (PUREX process) [42], dissolved in hydrochloric acid (HCl) or a combination of HCl and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), or anodically dissolved in ammonium carbonate solution. If the scrap is unalloyed and unclad, purification steps above may be omitted.

Additionally, uranium metal scrap can be directly fluorinated using fluorine gas [43]:



Electrorefining is another method that can be used to recover uranium from metal scrap; however, it has not yet been demonstrated on a production scale. In this method, scrap metal is loaded into an anode basket and submerged in molten  $\text{LiCl-KCl}$ . U is oxidized to the +3 state, migrates to the cathode, and deposits as metal [44]. In this case purification and reduction to metal are performed in a single process.

### 7.1 Pyrophoricity Hazard

Uranium scrap processed for recycle presents as significant pyrophoric hazard (see Section 3.2).

Additionally, several of the dissolution processes could present a pyrophoricity hazard: nitric acid dissolution of metal (such as is used in the PUREX process) can go into thermal runaway if not closely controlled and produces hydrogen gas as a byproduct, while dissolution with  $\text{HCl}+\text{H}_2\text{O}_2$  generates a considerable amount of heat.

### 7.2 Chemical Hazards

The predominant hazard associated with scrap recycle is chemical due to the many chemical reactions and manipulations necessary to purify the material and obtain a form suitable for conversion to metal. While all acids used throughout the process pose a hazard, by far the most hazardous is HF in both the gaseous and aqueous forms. Gaseous HF poses an immediate, acute hazard to the lungs and corneas if personnel are exposed. Aqueous HF is a contact hazard; it causes deep but painless burns upon contact that can lead to tissue death and also affects calcium metabolism through the formation of calcium fluoride ( $\text{CaF}$ ) and can lead to cardiac arrest and death.

Beyond the immediate hazard to personnel, fluorine is highly corrosive to most metals [6]. Any corrosion to pipes may eventually result in a release.

### **7.3 Radiation Hazard**

Uranium, especially DU [15], NU [16], and LEU, poses a very minor radiation hazard as it is only weakly radioactive. However, DU does emit fairly intense  $\beta$  radiation; inhalation of particulates can result in deposition of  $\beta$ -emitters in the lungs [17]. Employing ALARA principles of increased distance and decreased exposure time reduces the radiation hazard presented by the fuel material.

### **7.4 Nuclear Criticality Hazard**

Because criticality is highly dependent on multiple factors, it is difficult to make an overarching assessment of criticality hazards without knowledge of specific working conditions. Facilities should be evaluated by a criticality safety analyst and criticality safety engineer to ensure safe practices.

Criticality hazard is negligible when using natural uranium [16] without a special moderator (e.g., heavy water). The main criticality risks occur after enrichment and are the unexpected precipitation or settling of uranium compounds and the use of vessels that provide a favorable configuration for criticality.

## 8.0 Storage

This section describes the hazards associated with storage of fuel, fines/scrap, and other waste products, with emphasis placed on fines/scrap storage. Fines and scrap may be stored in two different forms and will have different storage hazards: some fines/scrap will be recycled (see Section 2.0), while the rest will be treated as waste. Since fines/scrap for recycle will only be in a metallic form for a short period of time, they can be left in metal form; however, scrap or waste that will be stored for an extended period should be in a stable form (generally as an oxide). Fuel can be stored as either bare fuel or clad [33], but may require different storage conditions. Much of the information presented in this section is taken from the *Safety Analysis Report for the Radioactive Scrap and Waste Facility* [45], an evaluation of scrap/waste storage at a facility located at Idaho National Laboratory (INL).

Several unsuccessful or unfavorable methods have been explored to stabilize uranium scrap [46]:

- Recovery by melting—melting was only achieved in outer regions; fire risk was increased from friction during compaction of scrap pieces
- Alloying with aluminum—no uranium was measured in the Al matrix; only fresh scrap can be treated using this method
- Wet chemical oxidation (a LANL procedure in which sodium hypochlorite reacts with U to form UOH, then sodium thiosulfate reacts with UOH to form UO<sub>2</sub>)—significant amounts of liquid waste are generated
- Burning

Fines oxidation by calcination was found to be the most favorable method for scrap uranium stabilization [21] [46]. Calcination has been demonstrated to have a high throughput and simple operation; in addition, calcination rate is controllable by varying gas composition in the furnace [46].

### 8.1 Pyrophoricity Hazard

Uranium metal reacts violently with water [21]; therefore, fines are best stored in their stable oxide form (UO<sub>2</sub>) in a dry, inert atmosphere or submerged in dry oil [21].

If uranium is stored in water and dissolved oxygen is limited, H<sub>2</sub> gas can be formed [21] according to the reactions



UH<sub>3</sub> is also a pyrophoric compound [21] and hydrogen is combustible.

Large pieces of uranium present a minimal fire risk (see Section 1.1). Additionally, U-10wt% Mo [31] and U-Zr [47] oxidize more slowly than pure uranium.

Several hazard sources were identified in the Safety Analysis Report (SAR) prepared for the Radioactive Scrap and Waste Facility (RSWF) [45]:

- Flammable materials (hydrogen in liners generated from internal corrosion, reactive metal-water reaction [Na, NaK] and/or radiolysis, pyrophoric metals [U, UH<sub>3</sub>], acetylene gas for liner cutting, vehicle fuel) can lead to fire or explosion

## 8.2 Chemical Hazards

High nitrate concentration in liquid waste may require treatment prior to release [4].

Several hazard sources were identified in the SAR prepared for the RSWF [45]:

- Flammable materials (hydrogen in liners generated from internal corrosion, reactive metal-water reaction [Na, NaK] and/or radiolysis, pyrophoric metals [U, UH<sub>3</sub>], acetylene gas for liner cutting, vehicle fuel) can lead to fire or explosion causing release of hazardous materials
- Hazardous materials in waste (lead, reactive materials [Na, NaK], toxic materials in storage liners) present a potential for hazardous material release and exposure

## 8.3 Radiation Hazard

Liquid waste may require dilution if the U-235 content in suspended solids is above a given control limit, dictated by permit discharge limitations [4].

Several hazard sources were identified in the SAR prepared for the RSWF [45]:

- Kinetic energy (movement of casks or containers) can initiate cask or container damage leading to radioactive material release
- Potential energy (suspended loads) has a drop potential leading to radioactive material release
- Pyrophoric materials (radioactive waste, U, UH<sub>3</sub>) can initiate a radioactive material release
- Radioactive materials in casks/containers and storage liners present a potential for radioactive material release and/or direct radiation exposure
- Flammable materials (hydrogen in liners generated from internal corrosion, reactive metal-water reaction [Na, NaK] and/or radiolysis, pyrophoric metals [U, UH<sub>3</sub>], acetylene gas for liner cutting, vehicle fuel) can lead to fire or explosion causing release of radioactive material
- Natural phenomena (earthquake, extreme wind, flood, lightning, etc.) can initiate radioactive material release and/or direct radiation exposure
- External events (fire) can initiate radioactive material release

## 8.4 Nuclear Criticality Hazard

Several hazard sources were identified in the SAR prepared for the RSWF [45]:

- Kinetic energy (movement of casks or containers) can initiate cask or container damage leading to criticality

- Potential energy (suspended loads) has a drop potential leading to criticality
- Fissionable materials in storage liners present a criticality potential



## 9.0 Summary

According to a 1996 International Atomic Energy Agency (IAEA) report [48], more than 80% of previously reported incidents were attributable to deficiencies in design or equipment (30%), deficiencies of management or procedures (10%), and operator or worker errors (44%). Therefore, proper procedures, management, and training can help to decrease incident occurrence. Each of the steps identified and analyzed involve pyrophoricity, chemical, radiation, and criticality hazards. While these hazards could be detrimental or even dangerous to infrastructure and personnel, there are also several ways to mitigate the hazards. Additionally, hazard analysis performed by Moon et al. [49] for the Reference Engineering-scale Pyroprocess Facility (REPF) determined that hazardous events are unlikely to occur with their hazard mitigations in place. Identified hazards for the various fuel fabrication steps are summarized in Table 9.1 and are accompanied by recommendations for hazard mitigation.

**Table 9.1.** Summary of hazards associated with each step of the fuel fabrication process and recommendations for hazard mitigation.

Hazard	Hazard Mitigation Recommendation	Fuel Fabrication Step
Pyrophoricity	Store unstable oxide powder in closed metal containers [14]	Feedstock Preparation
	Employ adequate ventilation to prevent buildup of flammable or oxidizing gases	Feedstock Preparation
	Employ adequate airflow for heat removal from self-heating [10]	Feedstock Preparation from Used Fuel
	Operate under an inerted atmosphere [5]	Feedstock Preparation from Used Fuel (sodium) [10] Reduction to Metal (charge loading and unloading) [18] Alloying and Casting [24] [33] Machining and Thermomechanical Processing Element/Assembly Fabrication (sodium) [33]
	Store fines/chips/swarf under liquids [17]	Reduction to Metal Alloying and Casting Machining and Thermomechanical Processing
	Use solid [22], oxygen-free, high-conductivity copper for VIM coil [25]	Alloying and Casting
	Reuse casting heel to reduce scrap/waste [22]	Alloying and Casting
	Control speed and use coolant during machining to reduce heat created [17] [23]	Machining and Thermomechanical Processing
	Co-extrude fuel cores and cladding to reduce the quantity of fuel alloy chips generated and the amount of machining required [35]	Machining and Thermomechanical Processing Element/Assembly Fabrication
Chemical	Employ adequate ventilation or respiratory protection to prevent exposure to uranium	Feedstock Preparation [15] Reduction to Metal [15]

Hazard	Hazard Mitigation Recommendation	Fuel Fabrication Step
	fumes, oxide powder, and metal fines, as well as other chemicals used in the fuel fabrication process	Alloying and Casting Machining and Thermomechanical Processing [15] [17]
	Have calcium gluconate on hand to treat HF exposure	Feedstock Preparation Scrap Recycle
	Substitute NF <sub>3</sub> for gaseous HF or F <sub>2</sub> due to its lower chemical toxicity [9]	Feedstock Preparation Scrap Recycle
	Use personnel protection equipment (PPE) when working with chemicals as necessary (apron, chemical-resistant clothing, gloves, safety glasses, face shield) [5]	Feedstock Preparation Machining and Thermomechanical Processing Element/Assembly Fabrication Scrap Recycle
	Employ corrosion-resistant materials such as monel where necessary	Feedstock Preparation Reduction to Metal Scrap Recycle
Radiation	Use PPE when working with radioactive materials as necessary (lab coat, gloves, safety glasses, respirator) [5]	Feedstock Preparation Reduction to Metal Alloying and Casting Machining and Thermomechanical Processing
	ALARA precautions (limiting time exposed and increasing distance from source)	Feedstock Preparation Reduction to Metal Alloying and Casting Machining and Thermomechanical Processing Element/Assembly Fabrication Scrap Recycle
	Conduct operations in hot cells	Feedstock Preparation from Used Fuel [10] Alloying and Casting (TRU [2], melt refining [10]) Machining and Thermomechanical Processing (TRU [2])
	Employ adequate ventilation or respiratory protection to prevent exposure to uranium fumes, oxide powder, and metal fines	Feedstock Preparation [15] Reduction to Metal Alloying and Casting Machining and Thermomechanical Processing [15] [17] [5]
	Conduct operations in a glovebox	Alloying and Casting [24] [33] [2] Machining and Thermomechanical Processing [2] [5]
	Wipe fuel to remove surface contamination [4]	Alloying and Casting
Criticality	Use vessels that do not allow for favorable criticality geometries or conditions	Feedstock Preparation Reduction to Metal Alloying and Casting Scrap Recycle
	Limit the size of charges	Reduction to Metal [18]

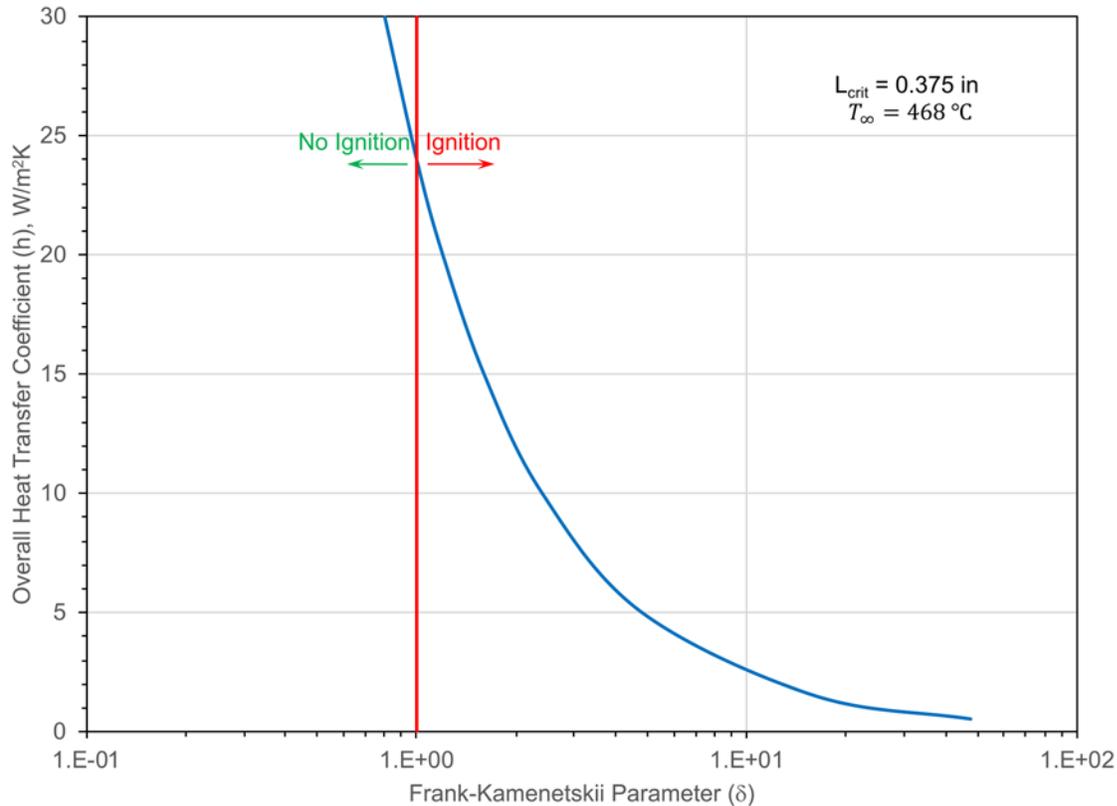
Hazard	Hazard Mitigation Recommendation	Fuel Fabrication Step
		Alloying and Casting [34]
	Avoid water cooling of induction coils [22] [25]	Alloying and Casting
Electrical	Clean fuel dust from furnace to avoid corona arcing [22]	Alloying and Casting
	Check induction coil for adequate coil spacing and problems with the coil insulator to prevent arcing [22]	Alloying and Casting
	Use appropriate coil material [22]	Alloying and Casting

The main hazard from uranium metal and uranium alloy fuel fabrication is pyrophoricity. Uranium is a highly reactive and pyrophoric material, and several steps of the fuel fabrication process can result in fines, chips, or powder. Several other materials, e.g., calcium and sodium, are pyrophoric as well and must be treated with caution.

The Frank-Kamenetskii parameter  $\delta$  can be used to determine the conditions for ignition:  $\delta \geq 1$  suggests ignition, with  $\delta$  defined as

$$\delta = \frac{V \cdot \Delta H \cdot C \cdot T_r}{h \cdot A \cdot T_\infty} e^{-T_r+1}, \quad (3.9)$$

where  $V$  is the volume,  $\Delta H$  is the effective heat of reaction per mass,  $C$  is a pre-exponential factor in the Arrhenius form of the material consumption rate,  $T_r$  is the ratio of the activation temperature  $T_E$  to the ambient temperature of the piece  $T_\infty$ ,  $h$  is the overall heat transfer coefficient, and  $A$  is the surface area [21]. A smaller piece of material (and therefore smaller  $A$  and  $V$ ) requires a higher ambient temperature for ignition. Also, use of coolant can increase  $h$  over the minimum value required to avoid conditions for ignition. Other variables are more or less constant for a given material system or process. **Figure 9.1** presents an example of the Frank-Kamenetskii parameter in practice for a reactive slab critical thickness of 0.375 inches and an ambient temperature of 468°C, taken from Burkes [21]. As can be seen in the figure, increasing the overall heat transfer coefficient above some threshold value changes the slab from ignitable to no ignition.



**Figure 9.1.** Overall heat transfer coefficient as a function of the Frank-Kamenetskii parameter for a reactive slab critical thickness of 0.375 inches and an ambient temperature of 468°C (from Burkes [21]).

The other considerable hazard is chemical. Acids and caustic chemicals are employed throughout the fuel fabrication process.

Radiation hazards mainly present as contamination concerns. Use of ventilation or gloveboxes can reduce or even eliminate risk of contamination by radioactive materials. Additionally, volatile fission products released during fuel reprocessing must be collected via traps in the exhaust system (i.e., between the melt furnace and vacuum pump used to evacuate the furnace) [10].

Criticality is influenced by several parameters, summarized by the acronym MERMAIDS. Because criticality is highly dependent on multiple factors, it is difficult to make an overarching assessment of criticality hazards without knowledge of specific working conditions. Criticality is only of minor concern as long as proper criticality safety as determined by analysts and engineers is observed. Historically, criticality accidents during fuel fabrication processes were generally a result of either unsafe geometry, reflection, or a combination of the two [12]. Most reported criticality accidents occurred between 1958 and 1964 when nuclear fuel processing was significantly scaled up without increasing criticality safety [1].

Canu et al. [11] evaluated occupational exposure to uranium and determined that it presented only a limited increase in cancer risk; however, further investigation is necessary due to low statistics and inaccurate quantification of internal exposure.

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## **Appendix A**

### **Table of References**



# **Appendix A**

## **Table of References**

The major information sources on uranium metal and alloy processing have been gathered to identify hazards associated with metal fuel preparation, processing, and fabrication. This section contains a table of these references, as well as the facility where work occurred, processes described, associated hazards, and events that have occurred for each reference.

**Table A.1.** Major uranium metal and alloy processing information sources, facility where work was performed, processes and hazards described, and events reported in the literature.

Document	Facility	Process Description	Hazards	Events
Bailey et al., “Fabrication of Aluminum-Plutonium Alloy Fuel Elements by Coextrusion”; HW-63151, December 1, 1959	Hanford	Fuel coextrusion	Contamination, chemicals	
Batte, G.L., “Fuels & Engineering Division (FED) Fabrication Processes”; January 6, 1992	EFL, FMF (INL)	Slug casting, element fabrication, subassembly fabrication	Sodium	
Bhanu et al., “Assay of uranium in scrap and waste produced at natural uranium metal production and fuel fabrication plants”; Journal of Radioanalytical and Nuclear Chemistry 121 (1988) 29-43	None	Extraction of uranium from various scrap and waste products	Waste, material holdup, chemicals	
Burkes, Doug, “RPL 48 Metal Cutting Operations Evaluation”; April 2018	PNNL		Fire, particulate release	
Burkes et al., “Metallic fast reactor fuel fabrication for the global nuclear energy partnership”; Journal of Nuclear Materials 392 (2009) 158-163	INL	Casting		
Burkes et al., “A U.S. perspective on fast reactor fuel fabrication technology and experience part I: metal fuels and assembly design”; Journal of Nuclear Materials 389 (2009) 458-469	EBR-II (INL) and FFTF (Hanford)	High level		
Canu et al., “Cancer risk in nuclear workers occupationally exposed to uranium—emphasis on internal exposure”; Health Physics 94 (2008)	Various		Worker exposure	
Durazzo et al., “Manufacturing low enriched uranium metal by magnesiothermic reduction of UF <sub>4</sub> ”; Annals of Nuclear Energy 110 (2017) 874-885	None	Magnesiothermic reduction of UF <sub>4</sub> to metal	Chemical, fire, dispersion	
Frajndlich et al., “Alternative route for UF <sub>6</sub> conversion towards UF <sub>4</sub> to produce metallic uranium”; Presented at The 1998 International Reduced Enrichment for Test Reactor Conference (1998)	None	Aqueous conversion of UF <sub>6</sub> to UF <sub>4</sub>	Chemical, pressure	

Document	Facility	Process Description	Hazards	Events
Gertsen, John, "UPF Capabilities Review"; "UPF Containment Requirements"; "UPF Technology Development"; OSD-CAPE Visit, April 9-10, 2013 [reference is UCNI]	UPF (Y-12)	Full process flow diagram (feedstock production, casting, forming, and machining)	Criticality, chemical, contamination, RADCON, personnel safety	
Hardy, C. J., "Possible trends and methods for the production of high-purity products"; AAEC Symposium on Uranium Processing	None	Dry and wet fluorination methods (feedstock and scrap recovery)		
Harrington et al., "Uranium Production Technology"; D. Van Nostrand Company, Inc. (1959)	None	Chemical purification, metal reduction, casting, extrusion, rolling, machining, scrap recycle	Chemical, criticality, waste, health hazard	
Hubbard et al., "U-10Mo Baseline Fuel Fabrication Process Description"; PNNL-26880 (2017)	Y-12, BWXT, Aerojet	Ingot production; fuel foil, fuel plate, and fuel assembly production	Machining, waste	
Jackson, Ross J., "Vacuum-induction melting, refining, and casting of uranium and its alloys"; RFP-4186 (1989)	Rocky Flats Plant	Vacuum induction melting/casting		
Jackson et al., "Extrusion of the uranium-0.75 weight percent titanium alloy"; RFP-2275 (1975)	Rocky Flats Plant	Casting, extrusion	Fire, machining	
Jessen, N. C., "Melting and casting uranium and uranium alloys"; Y/DV-213	Y-12	Casting, equipment requirements	Fire, contamination, electrical	
Kalsi et al., "Assay of uranium in U-bearing waste produced at Natural Uranium Metal Fuel Fabrication Plants by gamma-ray spectrometry"; Nuclear Instruments and Methods in Physics Research A 337 (1194) 594-597	None	Scrap and waste	Waste, material holdup	
Kang et al., "Oxidation behavior of U-10wt% Mo alloy in air at 473-773 K"; Journal of Nuclear Materials 304 (2002) 242-245	None	Metal oxidation	Fire	
Kim et al., "Fabrication of uranium alloy fuel slug for sodium-cooled fast reactor by injection casting"; Journal of Radioanalytical Nuclear Chemistry 301 (2014) 797-803	KAERI	Injection casting	Waste, electrical	
Kim et al., "Fabrication of U-10 wt.%Zr Metallic Fuel Rodlets for Irradiation Test in BOR-60 Fast Reactor"; Science and Technology of Nuclear Installations, vol 2016	KAERI	High level	Sodium	

Document	Facility	Process Description	Hazards	Events
Kittel et al., "History of fast reactor fuel development"; Journal of Nuclear Materials 204 (1993) 1-13	Various	High level		
Lee et al., "Metal Fuel Development and Verification for Prototype Generation IV Sodium-Cooled Fast Reactor"; Nuclear Engineering and Technology 48 (2016) 1096-1108	KAERI	High level		
Mariani et al., "Metallic Fuels: The EBR-II legacy and recent advances"; Procedia Chemistry 7 (2012) 513-520	FCF, EBR-II (INL)	Electrorefining	Fire, criticality	
Moon et al., "Identification of Safety Controls for Engineering-Scale Pyroprocess Facility"; Nuclear Engineering Technology 47 (2015) 915-923	REPF (KAERI)	Process flow diagram plus facility layout	Preliminary identification: spent fuel, radioactive materials, toxic materials, fire, electrical, ... (see Table 3)	
Morris, T.O., "Machining of Uranium and Uranium Alloys"; American Society for Metals Metallurgical Technology of Uranium and Uranium Alloys Seminar (Y/DX-372); May 26-28, 1981	Y-12	Machining, safety precautions	Fire, chemical, dust	
Operational Accidents and Radiation Exposure Experience within the AEC 1943-1975, WASH 1192 (Rev), 1975	Various			Fire, criticality, chemical
Processing and Applications for Depleted Uranium Alloy Products, Battelle Columbus Laboratories, AD-A033 938, September 1976 <a href="https://apps.dtic.mil/dtic/tr/fulltext/u2/a033938.pdf">https://apps.dtic.mil/dtic/tr/fulltext/u2/a033938.pdf</a>	None	Casting, machining	Fire, toxicity	
Ridal et al., "Stabilisation of Uranium and Thorium Metal Scrap"; Waste Management Symposia (1998)	ANSTO (Australia)	Calcination of scrap	Fire	
Safety Analysis Report for the Radioactive Scrap and Waste Facility (MFC-771); SAR-407 and TSR-407, 8/23/12	Radioactive Scrap and Waste Facility (INL)		Fire, criticality, pressure, chemical	
Significant incidents in nuclear fuel cycle facilities; IAEA-TECDOC-867, 1996	Various			Various (criticality, chemical, fire, etc.)

Document	Facility	Process Description	Hazards	Events
Stephen et al., “Criticality safety studies for plutonium-uranium metal fuel pin fabrication facility”; Annals of Nuclear Energy 53 (2013) 458-463	PUMP-F (India)		Criticality	
Uranium:properties, processes, controls. Watertown Arsenal Laboratories, Monograph Series 55, October 1962 <a href="https://apps.dtic.mil/dtic/tr/fulltext/u2/610614.pdf">https://apps.dtic.mil/dtic/tr/fulltext/u2/610614.pdf</a>	Watertown Arsenal Laboratories	Casting, hot working, cold working, machining	Fire, radiation exposure, waste	
Wachs et al., “Conceptual Process Description for the Manufacture of Low-Enriched Uranium-Molybdenum Fuel”; INL/EXT-08-13840 (February 2008)	INL	High level, Fuel Process Flow Diagram, facility footprint requirements	Chemical, radiological	
West et al., “Synthesis of Uranium Metal Using Laser-Initiated Reduction of Uranium Tetrafluoride by Calcium Metal”; LA-12996-MS (September 1995)	LANL	UF <sub>4</sub> reduction to metal	Chemical, laser, fire	
Wilkes et al., “EBR-II Fuel Slug Casting Experience”; ANL-IF-73, July 1987	EBR-II (INL)	Detailed description of casting process	Electrical, contamination, criticality, waste management	
Yahata et al., “Fluorination of Uranium Metal by Fluorine Gas”; Journal of Nuclear Science and Technology 9 (June 1972) 353-356	None	Scrap recovery by dry fluorination	Chemical, fire	
You et al., “Concept and safety studies of an integrated pyroprocess facility”; Nuclear Engineering and Design 241 (2011) 415-424	PRIDE (KAERI)	Process flow diagram plus facility layout	Radiation, electrical, chemical, fire, exposure	
ANSTO = Australia’s Nuclear Science and Technology Organisation; BWXT = BWX Technologies; EBR-II = Experimental Breeder Reactor-II; EFL = Experimental Fuels Laboratory; FCF = Fuel Conditioning Facility; FFTF = Fast Flux Test Facility; FMF = Fuel Manufacturing Facility; INL = Idaho National Laboratory; KAERI = Korea Atomic Energy Research Institute; LANL = Los Alamos National Laboratory; PNNL = Pacific Northwest National Laboratory; PRIDE = Pyroprocess-Integrated inactive Demonstration facility; PUMP-F = Plutonium-Uranium Metal fuel Pin fabrication Facility; RADCON = Radiation Control; REPF = Reference Engineering-scale Pyroprocess Facility; RPL = Radiological Processing Laboratory; UPF = Uranium Processing Facility				



## **Appendix B**

### **Key Terms Definitions**



# Appendix B

## Key Terms Definitions

Several key terms are used throughout this report that may not be familiar to readers. Definitions presented here are taken from Hubbard et al. [4] to provide a standard lexicon. All underlined words throughout the definitions are also defined here.

**aging** – a thermal process similar to annealing in which products are held at elevated temperatures for extended periods of time to allow precipitates to form, thereby strengthening the material. This can also be performed at room temperature with longer hold times

**alloying** – the addition of small amounts of other elements to a metal

**annealing** – a thermal process performed at elevated temperature to remove the effects of cold and hot working

**arc melting** – use of an electrode to perform a melt to create a master alloy

**can** – a case made of a sacrificial material to surround fuel elements during hot rolling and extrusion/co-extrusion, protecting against oxidation and radioactive contamination

**casting** – the process of pouring liquid metal into a mold to achieve a desired shape

**casting skull** – casting scrap that develops when liquid metal is not completely drained and freezes within the crucible

**cladding** – material that contains the fuel and fission products during operation

**co-rolling** – the process of bonding diffusion barrier or cladding to fuel during rolling

**cold rolling** – rolling performed at room temperature

**cold working** – a forming process performed at room temperature

**drawing** – a forming process in which material is pulled through a conical die, resulting in a reduction of cross-section. Typically performed at room temperature (cold working)

**electroplating** – the process of applying a coating with an applied voltage through the use of an anode (material to be applied to substrate) and cathode (substrate) submerged in an aqueous or salt solution

**extrusion** – a forming process in which a billet is pushed through a die to produce a rod, bar, seamless tube, or profile. Typically performed at elevated temperature (hot working)

**HIP can** – a metal container containing sandwiches of cladding and fuel with strongbacks between each sandwich to avoid bonding between cladding layers

**hot isostatic pressing (HIP)** – the process in which components are held under high pressure and temperature to achieve joining or to reduce porosity

**hot rolling** – a process using vertically opposed rolls or sets of rolls to reduce the thickness of a sheet or plate. Typically performed at elevated temperatures (hot working)

**hot top** – impurities in metal that float to the top during casting, which is cut off the casting piece and treated as scrap

**hot working** – a forming process performed at elevated temperature, including extrusion, hot rolling, etc.

**homogenization** – a thermal process similar to annealing in which products are held at elevated temperatures for extended periods of time to reduce segregation of alloying elements that occurred during casting

**master alloy** – feedstock added during casting that has been previously alloyed with elements to achieve the desired final composition

**physical vapor deposition (PVD)** – a coating process used to produce thin coatings (e.g., sputtering)

**pilgering** – a process typically used to form tubing by reducing the outer diameter with tapered rolls and the wall thickness with a mandrel located inside the tubing

**plasma spraying** – a thermal spray process that uses a plasma torch, working gas, and powder to propel molten material onto a substrate

**salt bath** – molten salt used to pre-heat material prior to hot working; the salt provides uniform heating and oxidation protection during heating

**strongback** – a solid piece of metal that is used to separate stacks of cladding and fuel during HIP

**swaging** – a forming process using dies to reduce the cross-section and produce the desired shape via hammering. Typically performed at room temperature (cold working)





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