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Investigation of the Airborne Release Fraction during Rapid Oxidation of Depleted Uranium Metal

by

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To the Graduate Faculty

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List of Abbreviations

μm – micron (1×10^{-6} meters)

AED – Aerodynamic Equivalent Diameter

AEGL – Acute Exposure Guideline Levels

AMAD – Aerodynamic Median Activity Diameter

ARF – Airborne Release Fraction

DCF – Dose Conversion Factor

DOE – U.S. Department of Energy

DP – DOE Office of Defense Programs

DU – Depleted Uranium

ERPGs – Emergency Planning Response Guidelines

FASB – Fuels and Applied Sciences Building

IAEA – International Atomic Energy Agency

INL – Idaho National Laboratory

m/s – meters per second

MDA – Minimum Detectable Activity

NIOSH – National Institute for Occupational Safety and Health

NRC – U.S. Nuclear Regulatory Commission

OSHA – Occupational Safety and Health Administration

PACs – Protective Action Criteria

ppm – parts per million

RF – Respirable Fraction

TEELs – Temporary Emergency Exposure Limits

TVL – Threshold Value Limit

Abstract

The Idaho National Laboratory (INL) performs research and development activities for the Department of Energy (DOE). Some research is to investigate new and alternative manufacturing methods for nuclear fuel fabrication. When metal-based (uranium) fuel fabrication techniques are investigated, the pyrophoricity of uranium metal is an inherent hazard. Therefore, a desire exists to understand the airborne release fraction (ARF) under the conditions of ignition (rapid oxidation), extinguishing and clean-up. To investigate the ARF, an experiment was designed in which 11 air samples were collected during six separate oxidation and cleanup trials for various depleted uranium (DU) masses, geometries and application methods for class D extinguishing media. An air sample was collected for each oxidation and cleanup evolution. Typically, one sample was collected during the oxidation and one sample was collected during clean-up giving two air samples per oxidation evolution. Health physics technicians (HPTs) ensured all experimental equipment was returned to a radiologically clean status before continuing to the next trial to reduce the potential of cross contamination. On the fourth trial only one air sample was collected due to a very rapid extinguishing of the oxidation process. ARFs were determined for each individual trial (oxidation and cleanup) as well as a total ARF (oxidation + cleanup). Prior to determining ARFs a comparative analysis between the alpha activities reported by the field measurements (thin window gross alpha/beta proportional counting) was made to the alpha spectroscopy results from the analytical laboratory. However, large percent differences were seen between the field measurements and analytical results making correlation between the two measurement techniques difficult and inconclusive due to minimal amounts of measurable activity on the air samples. Therefore, analytical alpha spectroscopy results were used exclusively for determination of the ARFs. In this research ARFs for DU never exceeded 1×10^{-5} for any single trial evolution and total ARFs fluctuated between a maximum of 7×10^{-6} to a minimum of 7×10^{-7} . The ARF values determined in this research effort agree well with early similar studies and are well below the extrapolated bounding ARF value 1×10^{-3} and median value 1×10^{-4} prescribed in DOE-HDBK-3010-94.

KEYWORDS: Airborne release fraction, uranium, depleted uranium.

1 Introduction

The Idaho National Laboratory (INL) performs research and development activities for the Department of Energy (DOE). Some of this research is to investigate new and alternative manufacturing methods for nuclear fuel fabrication. Metal-based (uranium) fuel fabrication techniques are investigated. An inherent hazard is the pyrophoricity of uranium metal, and it is therefore, desired, to understand the airborne release fraction (ARF) under the conditions of ignition (rapid oxidation), extinguishing and clean-up. The ARF is defined by the Department of Energy's Handbook, Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities as, "the coefficient used to estimate the amount of a radioactive material that can be suspended in air and made available for airborne transport under a specific set of induced physical stresses" (DOE, 2013).

The ARF is one of five factors used in estimating the airborne source term by a five-component linear equation. ARFs have typically been determined by empirical measurement of a specific material, such as uranium or plutonium, or by surrogate material subject to a stress under controlled conditions. The ARF is commonly used in conjunction with the respirable fraction (RF) which is the fraction of airborne radioactive material as particulate that can be transported through air and inhaled into the human respiratory system and is commonly assumed to include particles with sizes of about 10- μm aerodynamic equivalent diameter (AED) and less. Where the AED is the diameter of a sphere of density 1 g/cm^3 that exhibits the same terminal velocity as the particle in question (DOE, 2013).

1.1 Initiating Event

On February 8, 2017, an incident occurred in the Fuels and Applied Sciences Building (FASB) where un-sintered green pellets (pre-annealing/sintering) were inadvertently removed from a sealed container filled with inert gas. This resulted in the pellets reacting with air by rapid oxidation (C. R. Clark).

The pellets in this event were uranium silicide (U_3Si_2). Given the event happened during the fabrication process the exact dimensions of the pellets are unknown. However, previous finished pellet dimension have been of the following dimensions; $11.54 \pm 0.06 \text{ g/cm}^3$ which is approximately 94.5% of the theoretical density of 12.2 g/cm^3 . The pellets were pressed from aggregated powders of uranium and silicon with particle diameters less than $300 \text{ }\mu\text{m}$. The powder was pressed in a 9.525 mm (0.375 inch) diameter die. Final pellet dimensions for irradiation average $0.819 \pm 0.00037 \text{ cm}$ ($0.3225 \pm 0.00014 \text{ in.}$) in diameter with an average height of $0.612 \pm 0.022 \text{ cm}$ ($0.241 \pm 0.009 \text{ in.}$). (Harp, Lessing and Hoggan).

This event did not result in any personnel injuries or detectable spread of contamination. The event did serve to highlight that there is limited understanding surrounding rapid oxidation events and the phenomenon of the pyrophoric processes. This is expected since most of the comprehensive research and benchmark studies conducted in this area, particularly with uranium, was performed in the late 1950's through the early 1980's during a markedly different era of research and regulation. Those research efforts, during nuclear power's infancy and formative years are still the most widely cited references in this area of research.

1.2 Research Objective

As a result of the FASB event a controlled experiment was devised with the following three objectives. The third (3) objective is the focus of this research:

1. Training (visual documentation of the oxidation reactions)
2. A comparison of extinguishing methods for uranium metal fires, and
3. An opportunity to understand the ARFs associated with the oxidation process, cleanup process and the potential effects of different application methods of class D extinguishing media.

1.3 Hypothesis Testing

The following hypotheses will be tested:

Test 1: Depleted uranium (DU) oxidation process without extinguishing media applied:

H_{1,0}: There is not a difference in the ARFs between the oxidation process and the cleanup process ($H_{1,0}: \mu_{ox} - \mu_{cu} = 0$).

H_{1,A}: There is a difference in ARFs between the oxidation process and the cleanup process ($H_{1,A}: \mu_{ox} - \mu_{cu} \neq 0$).

The process (oxidation and cleanup) population means for the trials (1, 2, 3 & 6) where no extinguishing media was deployed will be assessed using the paired sample t-test with a cutoff value for determining statistical significance of $\alpha = 0.05$. A p-value of less than or equal to $\alpha = 0.05$ results in rejection of the null hypothesis. A p-value of greater than $\alpha = 0.05$ results in the null hypothesis not being rejected.

Test 2: DU oxidation process with extinguishing media applied:

H_{2,0}: There is not a difference in the ARFs between the oxidation process and the cleanup process ($H_{2,0}: \mu_{ox} - \mu_{cu} = 0$).

H_{2,A}: The highest ARFs occurs during the oxidation process ($H_{2,A}: \mu_{ox} - \mu_{cu} > 0$).

The process (oxidation and cleanup) population means for the trials where extinguishing media was deployed (Trial 4 & 5) will be assessed using the paired sample t-test with a cutoff value for determining statistical significance of $\alpha = 0.05$. A p-value of less than or equal to $\alpha = 0.05$ results in rejection of the null hypothesis. A p-value of greater than $\alpha = 0.05$ results in the null hypothesis not being rejected.

Test 3: Application of the extinguishing media affects the ARFs:

H_{3,0}: The application of extinguishing media does not affect the total ARF

$$(H_{3,0}: \mu_{w/o\ ext} - \mu_{w-ext} = 0).$$

H_{3,1}: The application of extinguishing media does affect the total ARF

$$(H_{3,0}: \mu_{w/o\ ext} - \mu_{w-ext} \neq 0).$$

The total ARF population means for the trials when no extinguishing media was deployed (Trial 1, 2, 3 & 6) and the total ARF population means for the trials when extinguishing media was deployed (Trial 4 & 5) will be assessed using the two-sample t-test with a cutoff value for determining statistical significance of $\alpha = 0.05$. A p-value of less than or equal to $\alpha = 0.05$ results in rejection of the null hypothesis. A p-value of greater than $\alpha = 0.05$ results in the null hypothesis not being rejected.

The air sample filter media was initially evaluated using the established INL standard field counting techniques. Subsequent alpha spectroscopy analysis was performed at GEL Laboratories LLC¹. The alpha activity was evaluated by direct comparison of the means of the alpha activity measured by the two measurement systems. Sample results obtained from GEL are decay corrected to the sample collection date and time, and background corrected when necessary. The reporting units for sample results, total error/uncertainty, and minimum detectable activity (MDA) are in dpm/sample for air filters (smears). The uncertainty is reported, per INL contract, at one-sigma combined standard uncertainty of the sample result (INL).

¹2040 Savage Road, Charleston SC 29417.

2 Literature Review

2.1 Uranium

Uranium, atomic number 92, is the heaviest naturally occurring element known and has atomic weight 238.02891 (J-Lab). Originally discovered by German chemist Martin Heinrich Klaproth (1789) in the material known as pitchblende in oxide form UO_2 , not in its pure form as he had originally believed. Uranium is named for the planet Uranus which had recently been discovered. The pure form of uranium was isolated by French chemist Eugene-Melchoir Peligot after noticing the previously believed ‘pure’ uranium was reacting oddly with uranium tetrachloride (UCl_4). In 1896 French physicist Antoine Henri Becquerel first discovered radioactivity from a sample of uranium (J-Lab).

The International Atomic Energy Agency (IAEA) defines uranium as a Low Specific Activity silver colored material in its pure form, similar to lead, cadmium and tungsten. Uranium is found in trace amounts in rocks and soil, in water and air, and in materials made from natural substances. Uranium compounds vary greatly in solubility. Uranium does form soluble compounds that may combine with other chemical elements and compounds that affect its mobility in and through the environment, as well as its toxicity. Some uranium forms are very inert and stay in the soil for thousands of years without moving downward into groundwater (IAEA, 2019).

Average concentrations of uranium in the soil is about 2 parts per million (ppm), which equates to approximately 2 grams of uranium per 1,000 kilograms of soil. Uranium concentrations in granite can range from 2 to 20 ppm. Higher concentrations (50 to 1,000 mg per kg of soil) are found in some soils associated with phosphate deposits. Uranium exists as dust in the air. The action of particle settling leads to dust deposition on the surfaces of water, plants, and soil hence it is commonly recycled within the biosphere. Most uranium in water is produced by rock/soil dissolution with only a small fraction from settling dust (IAEA, 2019).

2.1.1 Isotopes of Uranium

Uranium has 23 known isotopes that range in atomic weight from 217 to 242 (Baum, Ernesti and Knox). The three naturally occurring isotopes are ^{234}U , ^{235}U , and ^{238}U all are radioactive but only ^{235}U is fissionable (J-Lab). Table 2-1 describes the natural abundance and radioactive properties of the primary uranium isotopes (DOE, 2017) (ATSDR) (J-Lab).

Table 2-1 Natural Abundance and Radioactive Properties of Primary Uranium Isotopes

Isotope	Half-time (y)	Abundance		Decay Mode	Energy (MeV)	Yield (%)
		By weight %	By radioactivity %			
^{234}U	2.47×10^5	0.0055	49.03	Alpha	4.72	28
					4.77	72
^{235}U	7.1×10^8	0.72	2.27	Alpha	4.21	6
					4.37	17
					4.40	55
					4.60	5
^{238}U	4.51×10^9	99.28	48.70	Alpha	4.15	21
					4.20	79

Uranium continuously undergoes transformations through the decay process to ultimately become a stable element. This is a complex process involving numerous serial transformations resulting in a complex decay chain (DHHS 2013). There are two naturally occurring decay chains one is the uranium series, headed by ^{238}U , and the other is the actinium series led by ^{235}U . Figures 2-1 and 2-2 from DOE-STD-1136-2017 (2017) illustrate the two naturally occurring decay chains.

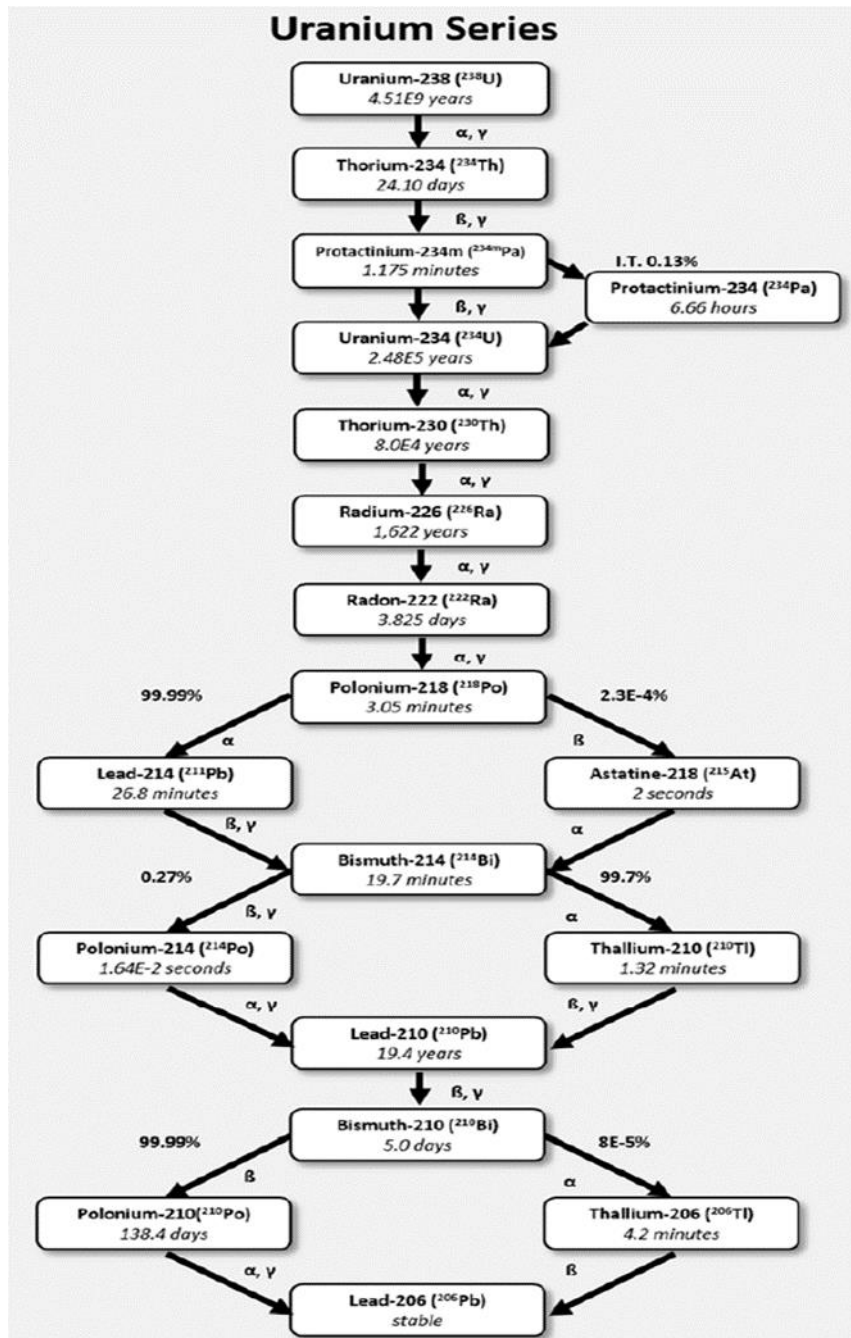


Figure 2-1 DOE-STD-1136-2017 uranium series

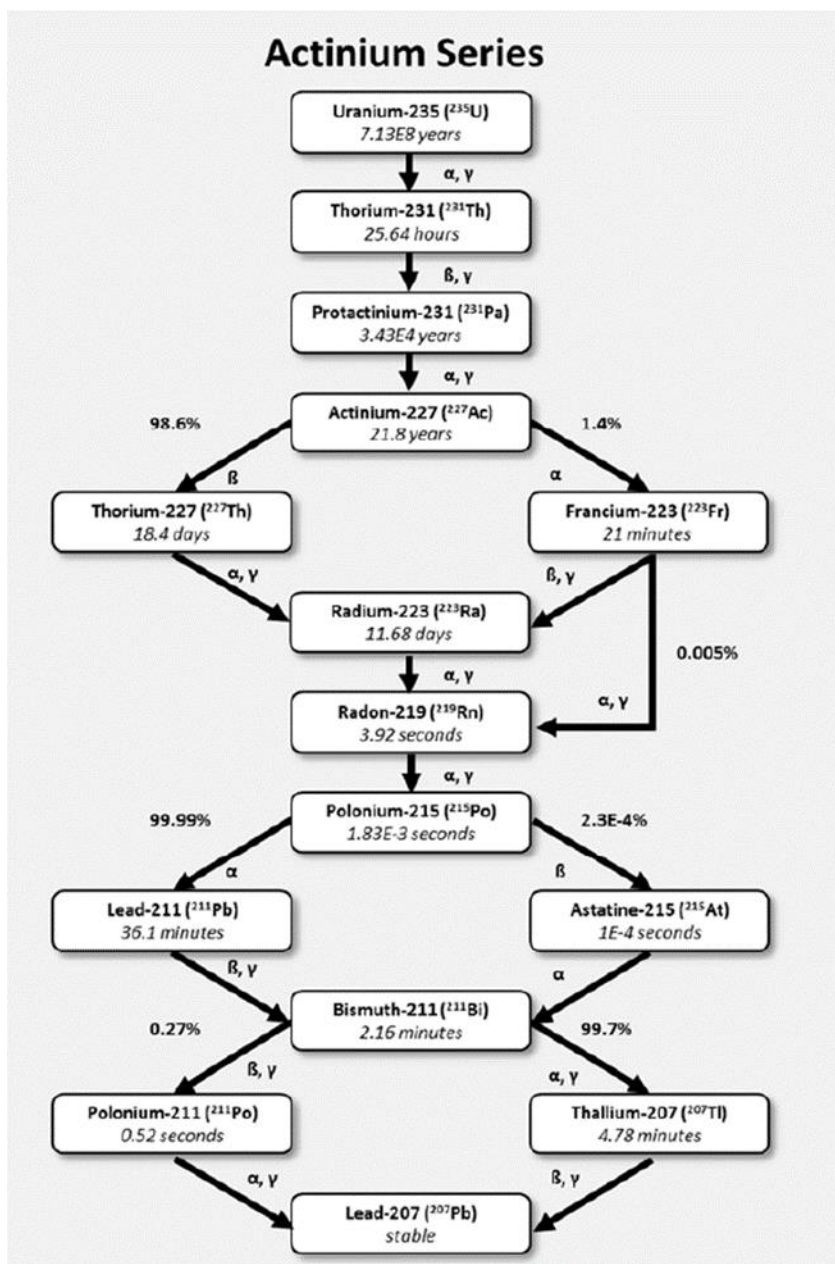


Figure 2-2 DOE-STD-1136-2017 actinium series

2.1.2 Depleted Uranium

Depleted uranium (DU) is uranium depleted in both ^{235}U and ^{234}U content as a by-product of an enrichment process (DOE, 2017). Typical isotopic abundance for depleted uranium are listed below in Table 2-2.

Table 2-2 Typical Isotopic Abundance of DU (g of Isotope per 100g of Material)

Isotope	Abundance By weight %	Specific Activity (Ci/g)
^{234}U	0.0007	
^{235}U	0.2	4×10^{-7}
^{238}U	99.8	

With its reduced radioactivity and high density, DU has many uses. Chief among them is radiation shielding, armor, counterweights, projectiles, and target elements in breeder reactors for plutonium production (DOE, 2017).

DU's physical and chemical properties have made it an excellent material for use in armor piercing munitions generally referred to a "kinetic penetrators". DU's high density, pyrophoric nature, and ability to self-sharpen by adiabatic shearing has given it a preferred status to other metals for military applications. Because military weapon systems go through extensive testing prior to deployment there is considerable information on the pyrophoric nature and subsequent ARFs available for review. Unfortunately, the use of DU munitions on the battlefields during the Gulf War and in Kosovo has provided population cohorts for determination of the health effects of DU on humans (IAEA, 2019).

2.2 Hazards of Uranium

With the advent of the nuclear age there has been widespread use of uranium involving mining of uranium ore, enrichment, and nuclear fuel fabrication in addition to the military applications of DU as previously stated. During the less regulated, formative years, of the industries involved in mining, milling, enrichment and fabrication of nuclear fuels smaller groups of the larger employee populations had unfortunate experiences through those many decades and now serve as population cohorts to further enhance our understanding of uranium hazards.

For miners the primary risk is not necessarily from uranium ore but from radon, specifically ^{222}Rn , which is one of many progeny of ^{238}U radioactive decay which happens to be a gas. The early uranium miners who worked in unknown, and often high, radon air concentrations demonstrated excess lung cancers and this risk of cancer appears to have increased with increasing exposure to radon. Studies of workers exposed to uranium in the nuclear fuel cycle have also been performed with reports of excess cancers, but no correlation with exposure has been seen, unlike the case for the miners (IAEA, 2019).

Generally, uranium is introduced into the body by ingestion or inhalation. A notable exception to this is veterans with DU shrapnel wounds. Both particle size and solubility determine the route and behavior in the body. Particles with an Aerodynamic Median Activity Diameter (AMAD) of five microns ($5\ \mu\text{m}$) are those of primary concern because of their ability to reach the deep alveolar region of the lung where they are dissolved and taken up by the blood (IAEA, 2019). Course particles that don't reach the deep lung will be removed by ciliary action and either ingested or expelled. About 10% of the more soluble uranium compounds absorbed by the blood through the lungs are transported to the kidneys where it tends to concentrate (IAEA, 2019).

While acute exposure to uranium compounds can cause nephrotoxicity in humans the evidence for similar toxicity as a result of long-term, lower-level occupational exposures is equivocal, given that epidemiological studies have not noted an increase in deaths from urogenital or renal diseases (ATSDR). Animal studies have shown the damage to the kidney was histologically manifested as glomerular and tubular wall degenerations. Ultrastructural analysis showed damage to the endothelial cells in the glomerulus, such as loss of cell process, and reduction in the density of endothelial fenestrae (Avasthi et al. 1980, Haley 1982; Haley et al. 1982; Kobayashi et al 1984). In the terminal segments of the proximal convoluted tubules, there was a loss of brush border, cellular vacuolization, and necrosis. Tubular reabsorption of solutes was disrupted. Functionally, this process led to a disruption of the tubular solute reabsorption and to a decrease in the filtration rate of the glomerulus, as assessed by creatinine or inulin clearance or by proteinuria (Bentley et al. 1985; Blantz 1975; Leach et al. 1973; Morrow et al. 1982a).

Excessive urinary excretion of protein, glucose, amino acids, or enzymes, such as catalase or alkaline phosphates are additional indicators of uranium-induced renal pathology (Maynard et al. 1953) by inhalation exposure (Bentley et al. 1985; Diamond et al. 1989; Haley et al. 1982; Leach et al. 1984; Maynard et al. 1953; Morrow et al. 1982a) (ATSDR).

This makes uranium, regardless of enrichment, a nephrotoxin and always a chemical toxicity hazard that must be considered. Most uranium ingested is excreted in the feces within a few days and never reaches the blood stream (IAEA, 2019).

According to the IAEA, “the general medical and scientific consensus is that in the case of high intake, uranium is likely to become a chemical toxicology problem before it is a radiological problem.” (IAEA, 2019)

2.2.1 Toxicity

A summary review of the toxic properties of uranium, chemical and radiological, is discussed in this section. The Agency for Toxic Substances and Disease Registry provides an excellent detailed summary document, “Toxicological Profile for Uranium” (2013) for the interested reader.

Early studies on the toxicity of uranium were performed in 1824 by Christian Gottlieb Gmelin. Several hundred experiments were performed prior to and during the development of the atomic bomb. With most efforts in recent years focusing on DU as a consequence of munitions used in modern conflicts such as Iraq and Kosovo (Kathren and Buklin).

Chemical toxicity is independent from enrichment and is, therefore, a function of the amount of uranium that can get into the blood, tissues and organs. The amount of intake is therefore directly related to the route of entry into the body and the solubility of the compound. Animal study data indicates relatively insoluble compounds of uranium such as UO_2 , U_3O_8 , and UF_4 have been shown to be nontoxic, even in acute doses, when given orally in large doses, up to 20% by weight in the diet, daily. More soluble compounds like UO_3 , UCl_4 , as examples, are toxic in large doses (2 to 10% in the diet), and less

soluble compounds like $\text{Na}_2\text{U}_2\text{O}_7$, $\text{UO}_3(\text{NO}_3)_2$, and UO_4 are toxic in moderate doses. As reported by Tannenbaum and Silverstone (1951), due to fluoride content in UO_2F_2 and UF_6 , following inhalation, the chemically toxic doses are even lower (Kathren and Buklin).

The specific activity of a compound is the primary determinant in assessing if chemical toxicity or radiotoxicity will be the controlling hazard. The radiotoxicity hazard is normally considered as a stochastic outcome (i.e., increased probability of carcinogenesis). Since natural uranium is a chemical toxin and classified as a “feeble poison” by Gmelin (1824) the radiotoxic hazard is only a consideration for uranium that has been enriched. Table 2-3 from DOE-STD-1136-2017 (2017) shows the controlling hazard based on exposure scenario, (acute vs chronic) solubility and provides the percent enrichment per solubility class where the change in controlling hazard occurs.

Table 2-3 Enrichments at which Radiological Limits are the Controlling Hazard (DOE-STD-1136-2017).

Material Type	Acute		Chronic	
	Using 100% of Radiological Limit	Using 2% of Radiological Limit (100 mrem)	Using 100% of Radiological Limit	Using 2% of Radiological Limit (100 mrem)
F	(a)	(a)	21.8% enriched	(b)
M	(a)	99.6% enriched	12.0% enriched	(b)
S	(a)	41.8% enriched	(b)	(b)

(a) Chemical toxicity limits are limiting at all enrichments.

(b) Radiological limits are limiting at all enrichments.

Rate of particle dissolution is divided into three categories by the ICRP Publication 68 model (1994b).

- Type F (fast): 10 min (100%)
- Type M (medium): 10 min (10%); 140 d (90%)
- Type S (slow): 10 min (0.1%); 7000 d (99.9%)

Types fast, medium and slow refer to the rate of absorption of materials in the pulmonary region of the lungs. However, many factors can affect dissolution rate therefore these general assignments should be regarded with caution (DOE, 2017).

Since determination of the controlling hazard is dependent on transportability (solubility in the body fluids), enrichment, and duration of exposure (acute or chronic) a comparative assessment is required for any suspected intake of uranium. This determination is typically, at least initially, performed based off field measurement data of the relative airborne concentration at a sampling point. For example, given a concentration a direct comparison can be made to the appropriate threshold value limit (TVLs) dependent on the regulatory agency. DOE recommends the more conservative of the two chemical toxicity standards (OSHA or ACGIH) be used to compare the chemical hazard with the radiological hazard. See Table 2-4 for a list of toxicological limits on airborne concentrations for uranium (DOE, 2017).

Table 2-4 Toxicological Limits on Airborne Concentrations of Transportable (soluble) Uranium

Agency	Chronic Exposure Occupational Limit, mg/m ³	Reference
NRC	0.2	Footnote to Appendix B, 10 CFR 20 (NRC, 1992b)
ACGIH ^(a)	0.2	<u>Threshold Limit Values and Biological Exposure Indices for 2005</u> , American Conference of Governmental Industrial Hygienists (ACGIH, 2005)
OSHA ^(b)	0.05 (soluble) 0.25 (insoluble)	29 CFR § 1910.1000
NIOSH	0.05	National Institute for Occupational Safety and Health (NIOSH)
(a) ACGIH also has a short-term exposure limit of 0.6 mg/m ³ . This is based on a 15-minute time weighted average exposure.		
(b) Preferred/recommended limit.		

The radiological dose can be determined by utilizing appropriate dose conversion factors (DCF) and models such as the ICRP Publication 68 (1994b) and ICRP Publication 78 (1997) which are illustrated in Table 2-5 (DOE, 2017).

Table 2-5 Dose Coefficients for Determining Enrichments above which Radiological Hazards Become Limiting

	Effective Dose Coeff, Sv/Bq	Effective Dose Coeff, Sv/Bq	Effective Dose Coeff, Sv/Bq	Organ Dose Coeff, Sv/Bq	Organ	Organ Dose Coeff, Sv/Bq	Organ	Organ Dose Coeff, Sv/Bq	Organ
	F	M	S	F		M		S	
U-234	6.4×10^{-7}	2.1×10^{-6}	6.8×10^{-6}	1.1×10^{-5}	BS	1.6×10^{-5}	LG	7.5×10^{-5}	ET
U-235	6.0×10^{-7}	1.8×10^{-6}	6.1×10^{-6}	1.1×10^{-5}	BS	1.4×10^{-5}	LG	6.9×10^{-5}	ET
U-238	5.8×10^{-7}	1.6×10^{-6}	5.7×10^{-6}	1.0×10^{-5}	BS	1.3×10^{-5}	LG	6.5×10^{-5}	ET
ET – Extrathoracic airways				LG – Lung			BS – Bone Surface		

Once the appropriate (effective or organ) dose has been determined based on solubility, enrichment and exposure the estimated dose should be compared to the appropriate radiological limit(s).

A complete treatment of the determination of the controlling hazard between chemical and radiological is beyond the scope of this research. However, the interested reader is encouraged to review DOE-STD-1136-2017 for illustrative examples of how to perform this determination.

In the case of acute chemical exposures under potential accident conditions protective action criteria (PAC) levels have been established by the DOE. Acute exposure guideline levels (AEGLs) represent threshold exposure limits for the general public and are applicable to emergency exposures ranging from 10 minutes to 8 hours. DOE guidance is to use the 1-hour AEGL values and those values are listed in Table 2-6 (DOE, 2018).

Table 2-6 Protective Action Criteria (PAC) Rev. 29a based on applicable 60-minute AEGLs, EPRGs, or TEELs. Values in mg/m³

Chemical Name	CASRN	PACs based on AEGLs, EPRGs, or TEELs (mg/m ³)		
		PAC-1	PAC-2	PAC-3
Uranium	7440-61-1	0.6	5	30
Uranium dioxide; (Uranium (IV) oxide)	1344-57-6	0.68	10	30
Uranium hexafluoride; (Uranium fluoride)	7783-81-5	3.6	9.6	36
Uranium hydride; (Uranium (III) hydride)	13598-56-6	0.61	5	30
Uranium oxide; (Triuranium octaoxide)	1344-59-8	0.71	10	50
Uranium telluride	12138-37-3	1.2	10	62
Uranium telluride (U ₃ Te ₄)	12040-21-0	1	8.5	51
Uranium trioxide	1344-58-7	0.045	0.5	3
Uranyl acetate; (Uranium oxyacetate)	541-09-3	0.98	5.5	33
Uranyl fluoride; (Uranium oxyfluoride)	13536-84-0	0.78	4.3	26
Uranyl hydroxide (includes liquids)	13470-18-3	0.77	4.3	26
Uranyl nitrate (solid); (Bis(nitrato-O,O')dioxouranium)	10102-06-4	0.99	5.5	33
Uranyl nitrate (yellow salt)	36478-76-9	0.99	5.5	33
Uranyl nitrate hexahydrate	13520-83-7	1.3	7	42
Uranyl nitrite (liquids)	z-0087	0.91	5	30

2.3 Uranium Chemistry

As reported by Lide (2008) uranium can exist in five oxidation states +2, +3, +4, +5, and +6; however, only the +3 and +6 states are of practical importance. The tetravalent (+3) oxidation state of uranium is reasonably stable and forms compounds of; hydroxides, hydrated fluorides, and phosphates. The hexavalent (+6) oxidation state is the most stable and forms the most commonly occurring compound U_3O_8 . Other major compounds for uranium include oxides, fluorides, carbides, nitrates, chlorides, acetates, and more. With the unique characteristic of UO^{+2} ions being fluorescent under ultraviolet light (ATSDR). Table 2-7 from DOE-STD-1136-2017 (2017) lists commonly encountered uranium compounds.

Table 2-7 Common Uranium Compounds, Chemical Name and Solubility Class (DOE-STD-1136-2017).

Uranium Compound	Chemical Name	Material Type (Inhalation Solubility Class)
Uranium hexafluoride	UF_6	Type "F"
Uranyl fluoride	UO_2F_2	Type "F"
Uranyl nitrate	$UO_2(NO_3)_2$	Type "F"
Uranyl acetate	$UO_2(C_2H_3O_2)_2$	Type "F"
Uranyl chloride	UO_2Cl_2	Type "F"
Uranyl sulfate	UO_2SO_4	Type "F"
Uranium trioxide	UO_3	Type "M"
Uranium tetrafluoride	UF_4	Type "M"
Uranium oxide	U_3O_8	Type "S"(b)
Uranium dioxide	UO_2	Type "S"(b)
Ammonium diuranate	$(NH_4)_2 + U_2O_7$	Type "M"(a)
Uranium aluminide	UAl_x	Type "S"
Uranium carbide	UC_2	Type "S"
Uranium-zirconium alloy	UZr	Type "S"
High-fired uranium dioxide	UO_2	Type "S"(b)

(a) Ammonium diuranate is known to contain uranium as UO_3 , and should not be assigned to a single inhalation class.

(b) The solubility of uranium oxides is very dependent on heat treatment. The rate of oxidation may also affect the solubility. It is recommended that solubility studies be performed to characterize the actual materials present.

The compounds listed above are considered by DOE-STD-1136-2017 (2017) to serve as general guidance only and recommends that transportability be independently determined by individual facilities utilizing accepted techniques for solubility determination (DOE, 2017).

2.3.1 Biokinetics

There is a large and comprehensive history of study on the biokinetics of uranium from both animal and humane cases involving both acute and chronic exposures. A concise summary is reported in NCRP Report No. 161, “Management of Persons Contaminated with Radionuclides: Handbook”. A summary of the committee report NCRP Report No. 161 is detailed below, as a complete treatment of the biokinetics of uranium is beyond the scope this project.

2.3.1.1 Systemic Biokinetics

Uranium is an actinide metal, but its skeletal behavior more closely resembles that of an alkaline earth element than those behaviors of other actinide elements like plutonium and americium according to Durbin (2006). The initial distribution of the uranium among different bones and bone parts is like that of calcium and there appears to be an exchange with Ca^{+2} and uranyl ions at the bone mineral surface. In relatively short periods after IV injection uranium becomes diffusely distributed in bone volume like calcium. This is the basis behind the ICRP (1995) adoption of the generic model structure for calcium-like elements being applied to uranium and is the same model used by NCRP Report No. 161. The parameter values illustrated in Table 2-8 are from the ICRP uranium model and the same used in NCRP Report No. 161. The values were based on several different sources of information including:

- measurements of uranium in blood and excreta of several humans who were intravenously injected with uranium;
- post-mortem measurements of uranium in tissues of some of those subjects;
- post-mortem measurements of uranium in tissues of occupationally and environmentally exposed subjects;

- data on baboons, dogs, or smaller laboratory animals exposed to uranium for experimental purposes; and
- consideration of the physiological process thought to determine retention and translocation of uranium in the body as reported by ICRP (1995) and Leggett (1994) (NCRP, 2009).

Table 2-8 NCRP Report No. 161 Transfer Coefficients in the Biokinetic Model for Uranium

Path	Transfer Coefficient (d⁻¹)
<i>Form plasma to:</i>	
Soft Tissue	1.05×10^1
Red-blood cells	2.45×10^{-1}
Urinary bladder contents	1.543×10^1
Kidney 1	2.94×10^0
Kidney2	1.22×10^{-2}
Upper large-intestine contents	1.22×10^{-1}
Liver 1	3.67×10^{-1}
Soft Tissue 1	1.63×10^0
Soft Tissue 2	7.35×10^{-2}
Trabecular bone surfaces	2.04×10^0
Cortical bone surfaces	1.63×10^0
<i>To plasma from:</i>	
Soft Tissue 0	8.32×10^0
Red-blood cells	3.47×10^{-1}
Kidney 2	3.80×10^{-4}
Liver 1	9.20×10^{-2}
Liver 2	1.90×10^{-4}
Soft Tissue 1	3.47×10^{-2}
Soft Tissue 2	1.90×10^{-5}
Bone surfaces ^a	6.93×10^{-2}
Nonexchangeable trabecular bone volume	4.93×10^{-4}
Nonexchangeable cortical bone volume	8.21×10^{-5}
From Kidney 1 to urinary bladder contents	9.90×10^{-2}
From Liver 1 to Liver 2	6.93×10^{-3}
From bone surfaces to exchangeable bone volume ^a	6.93×10^{-2}
From exchangeable bone volume to bone surfaces ^a	1.73×10^{-2}
From exchangeable bone volume to Nonexchangeable volume ^a	5.78×10^{-3}

^aApplies both to trabecular and cortical bone compartments.

2.3.1.2 Respiratory Tract Behavior

Behavior of inhaled uranium has, and continues to be, extensively studied. Laboratory animal studies along with accidental (acute) and chronic occupational exposures comprise the bulk of the available data that has been reported. Considerable variation in solubility has been observed in inhaled UO_2 and U_3O_8 with some studies indicating moderate solubility and others indicating low solubility in the lungs. As reported by Leggett *et al.*, (2005), data from urinalysis from a group of workers exposed to airborne uranium aluminide initially showed little dissolution but after several weeks in the lungs results showed rapid dissolution (NCRP, 2009).

DU oxides like those encountered in the Gulf War were mixtures of moderately soluble and relatively-insoluble materials as indicated by retrospective studies reported by Capstone (2009), DOD (2000), Guilmette *et al.*, (2005) and Parkhurst *et al.*, (2004). The chemical form for DU used in munitions seems to be dependent on the conditions of formation where particles formed by impacts may consist largely of U_3O_8 mixed with smaller quantities of UO_2 (NCRP, 2009).

2.3.1.3 GI Tract

Based on reviews by Legette and Harrison (1995) estimates of GI tract uptake of uranium in adult humans has been derived from the results of three (3) controlled experimental studies involving short-term oral intake of elevated quantities of uranium fluids from a controlled balance study performed in a metabolic research ward in a hospital and from numerous environmental studies in which urinary uranium was related to total intake or total excretion of the element in the same population. The controlled study demonstrated GI uptake values from less than 0.1 to approximately 6% for individual subjects, with central values for different studies in the range of 1 to 2.4%. Environmental studies estimated central ranges from 0.3 to 3.2%. This suggests that average GI uptake from ingested uranium expressed as a percentage of total intake of uranium from fluids and foods is approximately 1 to 1.5% for adult humans (NCRP, 2009).

2.3.1.4 Wound Behavior

The reader is referred to NCRP Report No. 156 for a complete and detailed review of the biokinetics of uranium in wounds. A substantial body of knowledge has been accumulated on the behavior of uranium in wounds based on a number of human exposure situations and a number of studies of implanted or injected uranium compounds in various species of laboratory animals (NCRP, 2009).

2.3.2 Uranium Metal

As a metal, uranium is combustible in finely divided forms and considered highly reactive and potentially pyrophoric. Given that, “finely divided” is an ambiguous term, for the purposes of this research, “finely divided” is considered any particle size less than 300 μm . This is consistent with the particle size of powder used in pellet fabrication in the FASB incident discussed in section one. Furthermore, the particle size range referenced by, “Industrial Ventilation, *A Manual of Recommended Practice for Design*, 29th edition” cites a particle size range from 0.001 to 100 μm for Metallurgical Dusts and Fumes (ACGIH).

Uranium’s radioactive properties do not influence its combustibility. Conversely, the radioactive properties are not influenced by burning metal. When handled in pieces, metallic uranium does not present a significant fire hazard unless exposed to severe and prolonged external flame but, once ignited the pieces burn very slowly. Unless strong drafts are present the combustion product, uranium oxide smoke, deposits in the immediate area of the burning metal (DOE, 2014).

2.3.2.1 Uranium Hydride

Uranium hydride (UH_3) is an important compound of uranium metal with respect to understanding the pyrophoric properties of uranium. The hydriding and subsequent dehydrating of uranium metal is a chief process whereby very pure but finely divided uranium metal is produced. The production of purified uranium metal takes place through serial processes of hydriding then dehydriding followed by subsequent sintering to produce a finely divided pure metallic form of uranium.

The formation of uranium hydride is a corrosion process where uranium metal is transformed from an elemental to a trivalent state and the hydrogen becomes chemically trapped at high elemental density.



The effects of the corrosion process by hydrogen not only produces UH_3 but results in the disintegration of the parent metal due to volume expansion as a result of the reaction. With enough hydrogen the reaction becomes self-propagating and occurs at a reaction rate four orders of magnitude higher than the rate of uranium oxidation under similar conditions (Banos, Harker and Scott).

The fundamental properties of UH_3 differ from the pre-cursor metal by a large change in density and resulting in a high surface area and appears as a finely divided black powder. Like its metallic counterpart UH_3 is highly reactive and pyrophoric under atmospheric conditions (Banos, Harker and Scott).

Particle size measurements of uranium powder by secondary electron (SE) images, as reported by Banos *et al.* (2018), showed individual particles appearing lozenge-shaped, or as elongated cuboids with a roughened exterior approximately $15 \pm 10 \mu\text{m}$ to $5 \pm 2 \mu\text{m}$. The reaction observed by Banos *et al.* (2018) is consistent with other descriptions given by investigators for uranium subjected to hydride-dehydride cycles. This suggests that corrosion of uranium metal produces a fine material that can potentially be easily dispersed (Banos, Harker and Scott).

The role of UH_3 in pyrophoricity was summarized by Smith (1956). When UH_3 is formed on the metal surface there is a likely potential the specimen will acquire an increase in pyrophoricity since the UH_3 is formed in an extremely finely divided state. This may be considered as the creation of small pieces of metal at the surface which helps to enhance the surface phenomenon to be discussed shortly. This process leads to an exceptionally large increase in the surface to volume ratio. In conjunction this process causes the replacement of the relatively non-porous UO_2 film layer at the metal surface with the

highly porous UH_3 providing increased susceptibility of the surface metal (newly exposed) to further attack by the surrounding atmosphere (e.g., oxygen, hydrogen, and/or water) (Smith). However, this is not the process that was used in this experiment as the powder used was put through the dehydriding process, so the resultant pellet contained only small amounts of residual UH_3 .

2.3.2.2 Uranium Oxide and Oxidation

Uranium metal's primary natural barrier against the external environment is an oxide layer that covers the surface. The principal oxides being those of UO_2 and UO_3 . Once formed these are known as a passivated layer. This layer decelerates further corrosion and helps to inhibit the onset of hydride formation (Banos, Harker and Scott).

As reported in a review of the published literature by Mishima, *et al.*, (1985) on the behavior of uranium in fire conditions, due to the similarity in matrix spacing, hyperstoichiometric uranium dioxide formed at the metal-atmosphere interface is adhering and limits the oxygen availability. At temperatures below 200 °C, the hyperstoichiometric dioxide, UO_{2+x} , is the principal product (DOE, 2013). This may be part of the reason for a lack of consensus in the literature when dealing with spontaneous ignition events as further highlighted in the following section.

When temperatures are slightly higher (between 200 to 275°C) a mixture of various other sub-oxides are reported to form (e.g., U_3O_7 , U_3O_8 , etc.) and at temperatures greater than 275 °C predominantly UO_2 and U_3O_8 are produced. At temperature between 350 °C and 600 °C the UO_2 formed is rapidly oxidized to U_3O_8 and falls away (spalls) as a fine black powder. From 650 °C to 850 °C the UO_2 forms a protective layer that at some point breaks away and at temperatures above 900 °C the UO_2 formed is adherent and protective. If water vapor is present in the air oxidation below 300 °C is accelerated and the same is true in carbon dioxide atmosphere up to 500 °C (DOE, 2013).

The presence of additives for phase-stabilization and alloying of uranium metal such as aluminum and titanium (staballoy) has the potential to change the first- or second-stage oxidation rates or break

weight or prevent transition to protective oxide formation that could result in a single accelerated oxidation rate (DOE, 2013).

Table 2-9 Potential Factors Influencing Uranium Oxidation (Table 4-9 - DOE-HDBK-3010-94)

Step	Potential Factors Influencing Rate
Metal surface of unit area oxidizing in air	1) Metal purity
	2) Metallurgical condition (grain size, strains, etc.)
	3) Temperature
	4) Time
	5) Gas composition
	6) Type of oxide film formed (protective or not)
Loss of heat of reaction by conduction to the surroundings	1) Thermal conductivity of metal
	2) Thermal conductivity of oxide coating
	3) Cross-sectional area at right angles to direction of heat flow
	4) Temperature gradient

2.3.2.3 Pyrophoric Properties of Uranium

Both finely divided uranium metal (U) and UH₃ are known to be likely to ignite at ambient temperature as was the case in the experiment at INL. However, there are several papers that report the lack of ignition of uranium hydride at room temperature and claim ignition is not achieved until temperatures reach 150 °C or more. In the experiments where ignition was not achieved at room temperature it is assumed that an oxide layer may have inhibited runaway oxidation (ignition). It was proposed by Ablitzer *et al.*, that at 140 °C, there is a change to the protective properties of the oxide layer that resulted in the uncovering of fresh hydride (Ablitzer, Le Guyadec and Raynal).

Uranium's pyrophoric properties were extensively investigated through the 1950s and 1960s. A sentinel report, "The Fire Properties of Metallic Uranium" by R.B. Smith, published in The Industrial Atom (1956) documents major insight into those properties. As reported by Smith (1956), the general fire properties of metallic uranium are like those of magnesium in similar form but are more pronounced than that of magnesium. Given the right conditions uranium is capable of self-sustaining combustion in air, nitrogen, oxygen, and carbon dioxide. Interactions with steam are far more energetic than those with

oxygen. Spontaneous, sometimes severe, fires have occurred with finely divided moist powders underwater and in inert atmospheres of helium and argon (Smith).

During self-sustaining combustion of uranium in air the emission of very intense but localized heat and light is typically observed while the rate of combustion can range from explosive, for powders dispersed in a cloud of dust, to very slow quiescent burning in the cases of massive metal (Smith).

Smith (1956) detailed some important points to understand before discussion of some of the reported vagaries experienced with uranium at time of publication:

1. Massive uranium is normally not capable of self-sustaining combustion in air without the continued application of heat from an external source.
2. The frequency of fires expected during machining operations with metallic uranium vary with type and rate of coolant applied.
3. Finely divided powder, no matter the method of fabrication, is particularly pyrophoric.

Smith (1956) summarized 13-separate incidents that demonstrated the pyrophoricity of metallic uranium varies over an extreme range of vagaries a selection considered most germane of those instances are listed below:

1. Incidental to the manufacture of metallic uranium powder, some of the powder was slowly accumulated in a sump under approximately 20 feet of sea water over the course of several months when without warning, a vigorous reaction took place resulting in a “geyser.” Following the eruptions, no further events occurred until subsequent buildup of metal powder resulted in another similar reaction.
2. Specimens of massive, low density (e.g., density of 15 g/cm^3) metallic uranium were reported to have spontaneously ignited in air after having rested without incident for several days on a metal shelf.

3. During the removal of the external jacket from failed fuel elements in a water-cooled reactor the existence of powder was observed, which was not present during fabrication. The powder spontaneously ignited when exposed to air.
4. Contrary to convention, large massive uranium pieces approximately one-half inch thick were able to achieve self-sustaining combustion after being exposed to heat from an external fire for approximately one hour. The ignited uranium continued to burn for nearly a day without the support of any external heat sources.
5. While conducting research with two small amounts of uranium powder, it was determined that they had been rendered “nonpyrophoric” after treatment with carbon tetrachloride. When repeated on a larger, third batch, of powder an explosion occurred resulting in severe injuries to the person conducting the experiments.
6. Uranium powders, typically prepared by pyrolysis of uranium hydride, are considered pyrophoric. However, the degree of pyrophoricity of such uranium powder is known to vary by a wide range.

Tentative conclusions reached by Smith (1956) based on his review of the events are:

1. Unusually high pyrophoricity may be acquired because of factors associated with the initial preparation of the metal. However, it may also be acquired at a later period by exposure to air and moisture under quiescent conditions.
2. Certain metal contaminants or stress in the metal (e.g., lathe chips) tends to increase the probability that a specimen will acquire increased pyrophoricity.
3. Spontaneous ignition of uranium involves rapid initial exothermic surface reaction with air or water. This contrasts with the better-known cases of spontaneous ignition (e.g., oily rags), of which uranium does not appear to follow, in which there is a gradual buildup of temperature.

4. The pyrophoricity of uranium is strongly influenced by the nature of the surface oxide. The oxides acquired during the metals exposure to dry air appear to be capable of reducing (if not eliminating) the ability of a specimen to acquire added pyrophoricity. If all other factors remain constant, the degree of pyrophoricity increases as the surface to mass ratio of the specimen increases.
5. Uranium combustion in air is promoted by the presence of moisture in limited amounts.
6. Low density metal specimens prepared by powder metallurgical means through decomposition of compressed uranium hydride is believed to have been involved in most if not all known fires in which massive pieces of metal spontaneously ignited under ambient conditions.

To explain pyrophoric instances detailed above Smith (1956) detailed what he termed, “particularly strong circumstantial evidence”:

1. It is almost invariably necessary for moisture to be present (especially in air) for larger particles of exposed metallic uranium to acquire unusually high pyrophoricity.
2. The probability increases greatly for a specimen to acquire unusually high pyrophoricity if it has been stressed and/or contains certain contaminants.
3. Unusually high pyrophoricity can exist as a purely surface phenomenon but may also be a property of all metal in the specimen.

Smith (1956) then summarized two instances in his report (beyond the scope of this work) where the effects of moisture greatly strengthen the circumstantial evidence:

1. Given suitable exposure conditions metal pyrophoricity can be increased by water.
2. No buildup of heat is need for spontaneous metal fires to occur.
3. There is most likely a connection between factors causing increase pyrophoricity and factors involved in water-metal explosions.

Previous work on uranium's pyrophoric and fire properties were conducted by Hartman *et al.* (1951) and supported what has been reported by Smith (1956). Hartman observed ignition of a 5-gram sample of uranium hydride powder under ambient conditions that closely resembled the observations of the INL experiment. A more recent study, "Uranium Pyrophoricity Phenomena and Prediction" Epstein *et al.*, (2000), supported the observations of Smith and Hartman. Other articles and reports reviewed by the author supported the general conclusions and observations concisely stated above by Smith (1956). A concise review of the pyrophoric properties of uranium is also reported in the DOE-HDBK-1081-2014, "Primer on Spontaneous Heating and Pyrophoricity" (2014) and is an excellent primary reference for novices in the field of pyrophoric materials.

2.4 Uranium Airborne Release Fractions

When reviewing the literature for references on airborne release fractions, one primary and comprehensive reference is DOE-HDBK-3010-94, "Airborne Release Fraction/Rates and Respirable Fractions for Nonreactor Nuclear Facilities," (1994, Reaffirmed 2013). This two-volume review and analysis of experimental data is the culmination of nearly four decades of investigation into the issue of airborne releases of radioactive material from nonreactor nuclear facilities. During four decades, starting in the 1960's, many individual experiments were conducted in the United States and other countries to develop data on release potentials. In the late 1970's the U.S. Nuclear Regulatory Commission (NRC) sponsored a research program to develop improved methods for realistically evaluating the consequences of major accidents in nuclear fuel cycle facilities culminating in NUREG-1320 (1988), "Nuclear Fuel Cycle Facility Accident Analysis Handbook." In the mid- to late-1980s DOE started placing increased emphasis on environmental, health, and safety issues and in response sponsored a program in the DOE Office of Defense Programs (DP) known as the Defense Programs Safety Survey (1993). One objective of the study was to build upon previous work to "develop consistent data and methodologies for making conservative estimates of basic consequence derivation parameters." Experimental data of airborne

release fractions and respirable fractions were summarized and evaluated to reasonably bound values for physical stresses associated with the experiments (DOE, 2013).

When reviewing the literature for airborne release fractions of uranium there are two benchmark studies that are continuously referenced. One performed by Carter and Stewart published in September 1970. The actual experiment write up was not found in the open literature and references made to the work in DOE-HDBK-3010-94 continuously uses the phrase, “reproduced from the source document,” to inform the reader that the information being provided was in too poor a quality to be directly copied into the text. The other benchmark study were sets of experiments performed by Elder and Tinkle and published in December 1980. Both sets of experimental data are heavily referenced throughout the open literature and are typically the comparative standard to which other experiments results are measured against. Mishima *et al.*, for example, characterized the oxide generated in the April 1983 burn test involving munitions with DU penetrators and performed a literature review on airborne release. The munitions were exposed to rigorous fire conditions in which 48 kg of DU (1-inch diameter by 30-inch long rods) were subjected to a wood and diesel fuel fire. The rounds were allowed to “cook off” and the DU was retained in the fire for a period of approximately three (3) hours with no detectable DU measured by air samplers surrounding the burn at distances less than 100 m. The oxides generated were collected and the particle size distribution, morphology and solubility in simulated interstitial lung fluid were measured. The fraction of the oxide produced in the burn that was associated with less than 10 μm aerodynamic equivalent diameter (AED) ranged from 0.2 to 0.65 wt per oxide (wt/o) and was predominately U_3O_8 which was originally classified as “Y” under ICRP 30 and would now be considered “S” under ICRP 66 (DOE, 2013).

Table 2-10 summarized the major results from the two benchmark studies for the combined ARF multiplied by the respirable fraction (RF). The respirable fraction is defined as the fraction of the ARF with particle size less than 10 μm AED.

Table 2-10 ARF x RF Results from Benchmark Experiments (from DOE-HDBK-3010-94)

Elder and Tinkle (December 1980)	
Parameters	ARF x RF
Air, up to 3.2 m/s, fire	5×10^{-3}
Air/Air-CO ₂ , 3.2 m/s, 500 °C	1×10^{-7}
Air/Air-CO ₂ , 3.2 m/s, 900 °C	4×10^{-6}
Carter and Stewart (September 1970)	
Parameters	ARF x RF
Air, static, molten metal	4×10^{-4}
Free-fall molten drops	6×10^{-3}

The Carter and Stewart experiments were performed to measure airborne uranium characteristics from molten metal under static (no movement metal) and dynamic (free-fall drop) conditions. Their mean and 95% confidence level ARF x RF from oxidation of static molten metal with airflow around and over the metal were 1.1×10^{-4} and 3.6×10^{-4} respectively (DOE, 2013).

The Elder and Tinkle experiments were performed on DU rods (kinetic penetrators) in armor defeating weapons. The rods were staballoy (beta-stabilized uranium, 99.25% uranium + 0.75% titanium) with a nominal diameter of 25.9 mm (about 1 inch), and a length of 0.345 m (13.6 inches) with a nominal weight of $3,355 \pm 3$ grams. The rods were heated in a rack in an up flow of air (heat + air or air-carbon dioxide atmosphere). For the first three tests, the heat was generated using either ignited uranium turnings or munition propellants. The fourth test used 10 batches of packing materials (wood and paper) for the heat source. Observations showed very little oxidation for the first three tests. However, 42% to 47% of the three rods in the fourth burn test were oxidized. This greatly exceeds the largest oxidations Elder and Tinkle (1980) had observed in their laboratory experiments and was attributed to the spalling of the oxide coat formed during the temperature fluctuations resulting from introduction of the 10 batches of fuel. The individual 5-min air samples taken during portions of four fuel additions showed apparent airborne concentrations ranging from 4.2 to 783 mg U/m³ with fractions < 10 µm AED from 20% to 62%. Unfortunately, the ARF determination from the experiments is difficult because the large variation in the airborne concentration and the times when the air samples were extracted are not reported (DOE, 2013).

Elder and Tinkle (1980) did measure the oxidation rates and airborne release for thirteen laboratory experiments in air of 50% air – 50% carbon dioxide at temperatures from 500 to 1,000 °C. For twelve of the tests, a gas velocity of 2.23 m/s (5 mph) was passed around the oxidizing rod. One experiment performed at 700 °C was done under static conditions (no gas flow). Under the laboratory conditions no self-sustaining reactions were observed and the oxidized fractions ranged from 6.0% to 30.2%. As previously discussed, only burn four with the wood and paper fuel in 10 batches exceeded the approximate 30% oxidation fraction observed in the laboratory. Total aerosol mass ranged widely with temperature and oxidizing atmosphere from approximately 1×10^{-4} for air only between 500 to 600 °C up to about 1×10^{-2} at 1,000 °C in a 50% air – 50% air-carbon dioxide atmosphere. The general trend shows an increase in total aerosol mass with increasing temperature. The mass of the particles less than 10- μ m AED has values that ranged from about 1×10^{-7} at 500 °C to 8×10^{-6} at 700 °C and 900 °C for the fraction of the total mass oxidized. When these values are adjusted, or extrapolated, to allow for complete oxidation of the metal the ARF values increase in range to 1×10^{-4} to 4×10^{-2} with RF from 0.006 to 0.17. The ARF x RF then ranges from 5×10^{-5} to 4×10^{-3} and are comparable with values reported by Carter and Stewart. The following conclusions are listed by the authors (Elder and Tinkle) (DOE, 2013):

1. Uranium particles in the respirable size range (10- μ m AED and less) were made airborne when the rods were exposed to temperature exceeding 500 °C for time greater than about 0.5 hours.
2. Production of oxide and airborne materials were enhanced by forced draft and temperature cycling during oxidation.
3. Metal rods with the test configurations did not exhibit any tendency towards self-sustained oxidation, although complete oxidation would, without doubt, be achieved if adequate fuel and time (greater than 4 hours) were provided (DOE, 2013).

DOE-HDBK-3010-94 continues to cite another literature review of DU munitions studies that have been performed which is beyond the scope of this project. The summary paragraph on uranium

oxidation at elevated temperatures for DOE-HDBK-3010-94 pulls together the results of the Carter and Stewart (1970) experiments as well as the Elder and Tinkle (1980) experiments and establishes the following ARF and RF values of 1×10^{-3} and 1.0 as the appropriate bounding values for thermal stress configuration. The solubility class is assumed to be class “S” based on the sintered oxides from the wood-oil fires involving DU rods in munitions and is concluded to also be a bounding solubility value (DOE, 2013).

Recently an effort to characterize respirable uranium aerosols from various uranium alloys in fire events was performed at the Y-12 National Security Complex by Clark (2015). His experiments indicate a distinct order of magnitude difference between uranium alloy responses to thermal stress. Clark (2015) details there is a minimal amount of the experimental data that exists on the formation and release of respirable oxide fumes from uranium alloys. He cites two primary guidance documents for quantifying the release of respirable oxides are the American National Standard for Airborne Release Fractions at Non-Reactor Nuclear Facilities (ANSI 1998) and the Department of Energy’s Handbook of Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities (DOE 1994). Both documents were principally authored by the late Dr. Jofu Mishima using the same benchmark studies of Carter and Stewart (1970) and Elder and Tinkle (1980) (D. K. Clark).

Clark details that the bounding ARF of 1×10^{-3} comes from the Elder and Tinkle (1980) experiments on beta-phase stabilized DU specimens of staballoy containing 0.75% titanium and the median ARF of 1×10^{-4} for uranium aerosol results from Carter and Stewart (1970) on alpha-phase uranium. The order of magnitude difference in the two values is indicative of a potential difference in alloy-specific responses to thermal stress (D. K. Clark).

The experiments performed by Clark (2015) were designed to mirror those of Carter and Stewart (1970) and Elder and Tinkle (1980) as closely as practical given the differences in the experimental environments from the 1970s and 1980s to the present. Clark (2015) results determined that the thermal response of U – Ti is significantly higher than the thermal response of other uranium alloys. As such,

RFs based on that alloy are not representative of pure uranium or other alloys. The Elder and Tinkle (1980) data used to support the ARF x RF of 1×10^{-3} in DOE-HDBK-3010-94 is specific to U – Ti alloy and the Carter and Stewart (1970) data supporting the median ARF x RF of 1×10^{-4} in the DOE guidance is appropriate for pure uranium and U – Nb alloys (D. K. Clark).

3 Experiment Setup

The experiment was performed by exposing a compacted pellet of fine uranium powder to ambient atmosphere in a controlled manner. The pellets were pressed right-cylinder compacts of uranium metal powder. The powder was made via the hydride-dehydride process. The powder production and pressing were performed in an inert atmosphere glovebox. The pellet(s) were then individually loaded into a sealed crucible, seen in Figure 3-1, made from solid stainless steel.



Figure 3-1 Reaction Crucible.

An assembled crucible is at right and at left is the open crucible showing the convex surface. As seen in the figure the crucible(s) have an O-ring seal and convex interiors that kept the pellet centered in the crucible. The loaded crucible was transferred out of the glovebox and carried to a radiologically controlled fume hood where a stage, shown in Figure 3-2, was employed to thermally isolate the experiment from the fume hood.

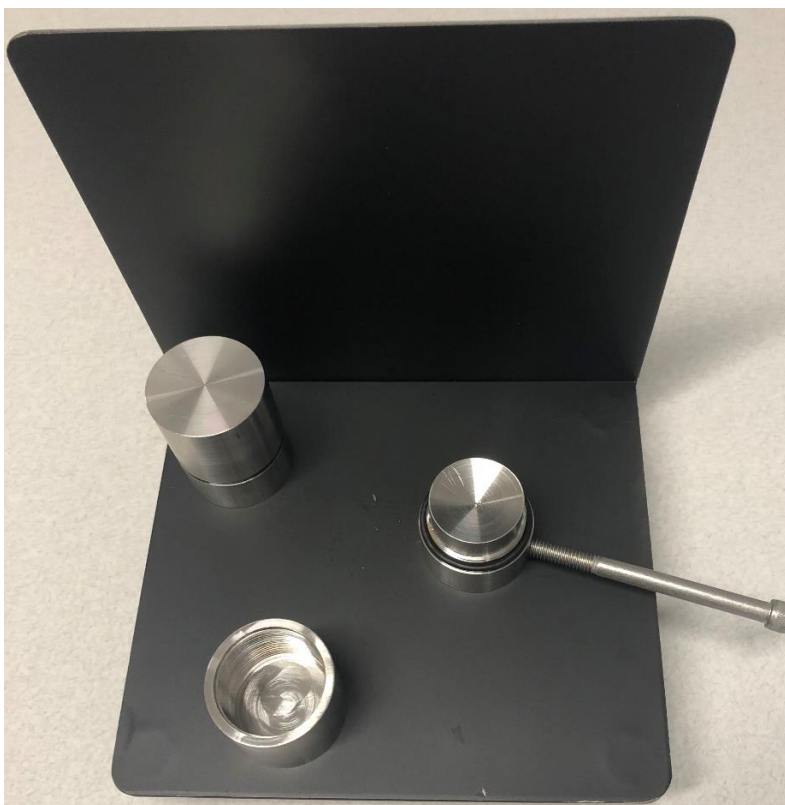


Figure 3-2 Reaction stage with crucibles

The airflow into the hood was checked to ensure the stage did not interfere with the fume hood's primary function to provide contamination control and confinement. Once in the fume hood, the lid was removed from the crucible and the oxidation process was observed to start nearly instantaneously. A total of six separate oxidation and subsequent cleanup trials were conducted as detailed in Table 3-1.

Table 3-1 Oxidation Trials

Pellet ID No.	Powder Weight (g)	Pellet Weight (g)	Pressing Conditions (N/m² + duration)	Conditions Assessed
CB1	4.001	3.987	1.38x10 ⁷ N/m ² for 30 sec	Primary ignition of pellet.
CB2	4.001	3.989	1.39x10 ⁷ N/m ² for 30 sec	Secondary ignition of DU block (block below pellet)
CB3	4.001	3.989	1.52x10 ⁷ N/m ² for 30 sec	Secondary ignition of DU block (block above pellet)
CB4	4.004	3.992	1.39x10 ⁷ N/m ² for 30 sec	Class D media applied by bag
CB5	4.001	3.994	1.43x10 ⁷ N/m ² for 30 sec	Class D media applied by shaker can
CB6	4.004	3.990	2.65x10 ⁷ N/m ² for 30 sec	Primary ignition of higher density pellet

The powder weight mentioned in Table 3-1 is the mass of DU powder via the hydride-dehydride process and the pellet weight is the subsequent weight of the pellet after compaction. There is a small (between 7 to 14 mg) reduction in mass when the powder is compacted into the pellet due to normal fabrication losses. Fractions of material are lost at each phase of fabrication by several ways. For example; transport losses when moving material, contact loss on the surfaces of handling and process equipment, as well as measurement repeatability and human performance factors.

Tests that considered the behavior of bulk materials were performed to investigate secondary ignition of the bulk DU during the oxidation of the compact material. The apparatus employed for this part of the investigation was a uranium block with a hole sized to fit the compact pellet. The uranium blocks, complete with a compact pellet inserted into its pre-drilled hole, were loaded into the crucible in order to conduct these tests.

Conservative bounding calculations of the energy release and temperature rise in the crucible were performed by the experiment staff. It was assumed that all the uranium would be converted to U_3O_8 per the following reaction:



U_3O_8 was chosen as a conservative bounding case since the molecule has the largest heat of formation of the oxides.

It was also assumed that all the heat generated would stay in the crucible and the actual mass of the pellet and bulk uranium would be under the masses used for the calculations (5 and 10 grams, respectively). Based upon these conservative assumptions, it was determined that for the pellet and the bulk DU oxidation a maximum of 75 kilojoules could potentially be released. The corresponding temperature rise was calculated to be 397 °C, well below the 1,400 °C melting temperature of stainless steel (Clark and Krawczyk).

During six (6) separate oxidation and cleanup trials a total of 11-air samples were acquired. An air sample was collected for each trial. One sample during the oxidation process and one sample during

cleanup operations for a total of two (2) air samples per oxidation and cleanup evolution. Health physics technicians (HPTs) ensured all experimental equipment was returned to a radiologically clean status before continuing to the next trial to reduce the potential of cross contamination. On the fourth trial only one air sample was collected due to the very rapid extinguishing of the oxidation process after the application of class D fire extinguishing media. The data in Tables 4-1 and 4-2 provide the field evaluation of the air samples relative to these tests.

3.1 DU Pellet Fabrication

Pellet fabrication was a three-step process of cleaning, hydride-dehydride, and compaction. Cleaning/etching the parent DU by immersing the material in a nitric acid bath (HNO_3) and spot cleaning with a nitrate acid-soaked pad/wipe (Moore, Fielding and King).

The cleaned DU was cycled through a hydride-dehydride apparatus. The process was initiated by heating a uranium alloy ingot in a hydrogen atmosphere. At moderate temperatures (typically $< 300\text{ }^\circ\text{C}$) the uranium reacts with hydrogen. The uranium hydride has a much lower density than the uranium metal ($10.9\text{ vs }19\text{ g/cm}^3$) and it sloughs off the ingot surface as hydride powder. After the Hydriding step is completed, the uranium hydride is reduced by heating the hydride powder under vacuum. The hydrogen slowly dissociates from the powder leaving only the uranium alloy (DU in this case) in powder form (Clark, Meyer and Strauss).

To complete the process, the finely divided and dehydrided DU powder was compacted per the parameters detailed in Table 3-1. The application of pressure to the powder increases the bulk density resulting in a solid compact pellet (Harp, Moore and Benson).

The resultant compact pellet(s) averaged just under four (4) grams and were compacted in a 1.0 cm diameter die. Given the timetable of the experiment, precise geometry measurements were not taken. The aim of the pellet fabrication was to hit the target mass of four grams and therefore no height measurements were taken on any of the pellets. Post experiment estimations, performed by the research

staff, have estimated the pellet density at approximately 30 to 35% of the theoretical density (for pellets CB1 through CB5) based off visual examination of the experiment video(s).

3.2 Oxidation

Unfortunately, no heat or heat production rate measurements were successfully recorded during the experiment. Figure 3-3 is the oxidation reaction in the first 2 seconds after the crucible lid was removed for the first pellet. Figure 3-4 is the pellet prior to spalling. There is an observable reduction in the visible light and heat output of the reaction as the available unreacted surface area retreats to the interior of the pellet slowing the reaction. In Figure 3-5 spalling of the pellet has occurred and the oxidation reaction continues in the newly exposed interior of the pellet albeit at a slower rate. Figure 3-6 shows the remnant of the completely oxidized and disintegrated pellet.



Figure 3-3 Oxidation Reaction. Approximately 2 seconds after Crucible Lid Removal

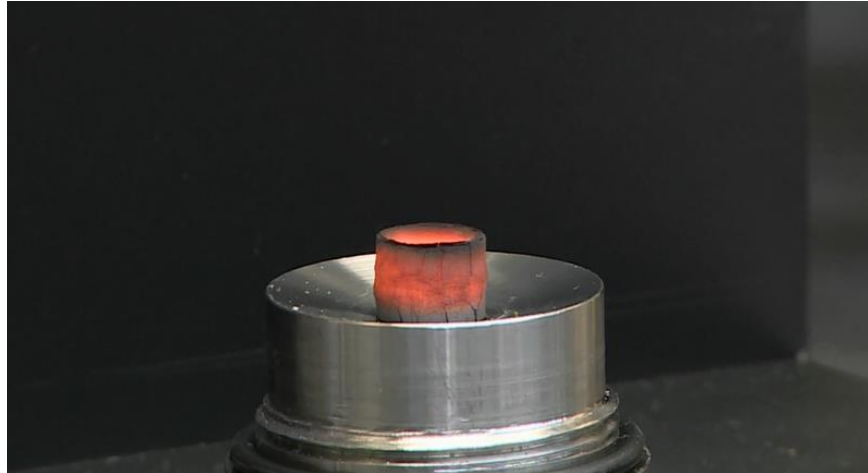


Figure 3-4 Oxidation Reaction. Prior to Spalling



Figure 3-5 Post Spalling Continued Oxidation



Figure 3-6 Complete Oxidation

3.3 Air Sampling

Air sampling equipment used during the experiment consisted of a giraffe/goose neck air sampler powered by a quarter horse power AC Marathon electric motor with a HI-Q air sampling head (P/N: RVPH-25) a five-micron Fluoropore 47-mm filter (Millipore). Samples were obtained at a flow rate of 99.11-liters per minute (3.5-cubic feet per minute) as measured by a HI-Q flow rate meter. Figure 3-7 illustrates the configuration of the air sampling equipment used, however, it is not the actual equipment.

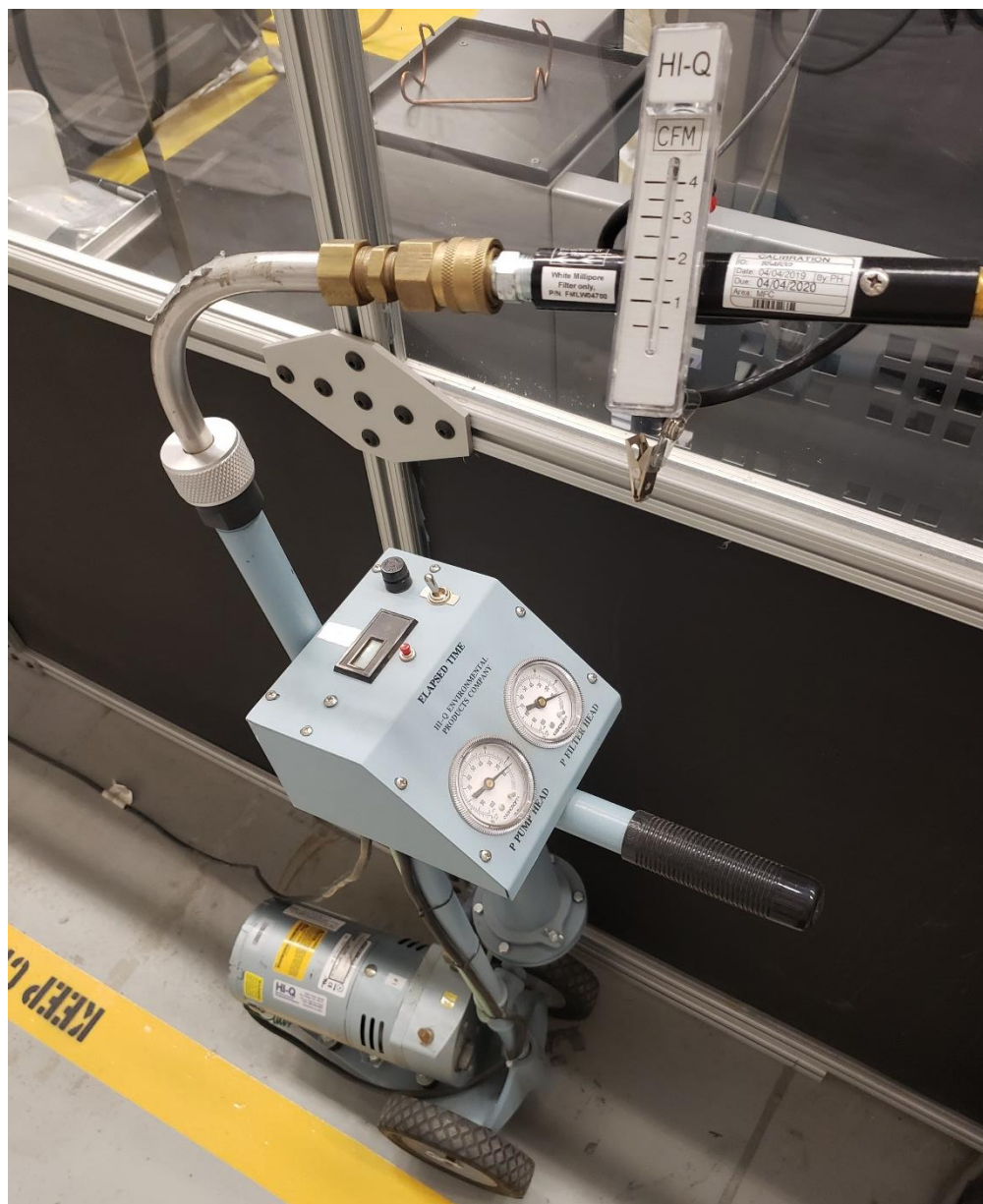


Figure 3-7 Air Sampling Equipment

The sample head was connected to the giraffe air sampler by 1.27-cm (0.5-inch) inside diameter (ID) (0.239 cm (0.094-inch wall thickness)) Tygon® tubing of approximately 1.5 meters (5 feet) in length and was located inside the fume hood approximately 46-cm (18-inches) above the reaction stage. The sampling head was in an optimized location to ensure it would not interfere with the experimenter but still be in the airflow path. The airflow path was verified using visual smoke (small area smoke test) prior to

oxidation of the pellets. Figure 3-8 illustrates the location of the sampling head inside the fume hood in relation to the reaction stage and crucible.

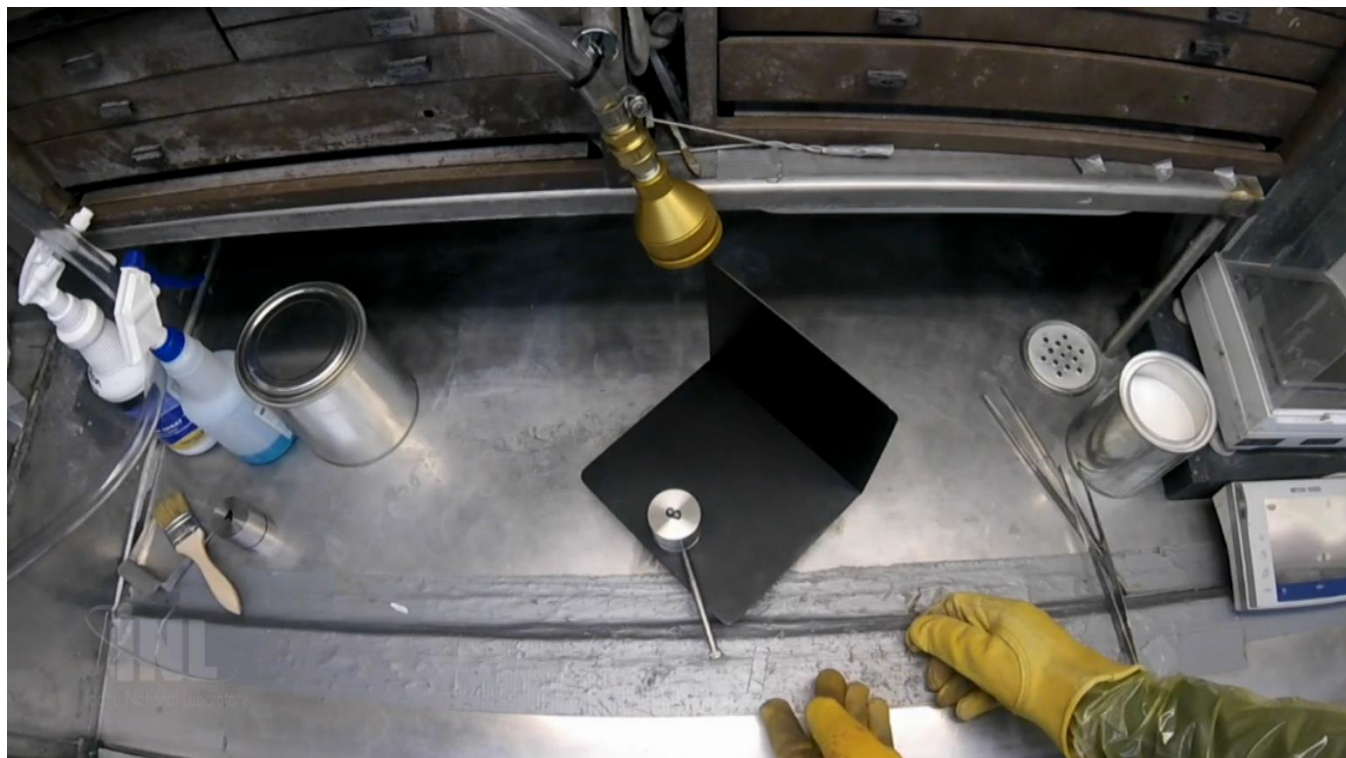


Figure 3-8 Air Sample Placement

4 Results and Discussion

The radioactive materials collected on air samples during the experiment were quantified by two methods (1) in the field per standard INL radiological controls air sampling procedures and (2) the same air sample filters were sent to an analytical laboratory for activity determination by alpha spectroscopy. No secondary ignition was observed in either trial where bulk uranium blocks were used.

4.1 Field Measurements

As is standard practice with air samples, a seven to ten day wait period was used post experiment to account for environmental interference in the activity measurements from naturally occurring environmental radioactive materials (i.e., radon and thoron progeny).

Air samples were counted on a Canberra Series 5 XLB Automatic Alpha Beta Counter which is a low background alpha/beta counter that utilizes a gas flow proportional detector. The counter is an automatic single detector, ultra-low background counting system. The instrument uses the Gas Stat digital gas conservation and monitoring system. The system has a high performance 5.7-cm (2.25 inch) gas-flow detector with ultra-thin 80- $\mu\text{g}/\text{cm}^2$ window and an advanced electronic diagnostics that continuously monitors operating conditions (INL, 2015).

Tables 4-1 and 4-2 summarize the counting data from the air samples collected by the HPTs during the experiment trials.

Table 4-1 Oxidation Process Air Samples

Air Sample ID	Pellet ID	Sample Run Time (min)	Total Volume Liters (ft³)	Alpha Activity (dpm)
1B	CB1	27	2,676 (94.5)	1.0
2B	CB2	13	1,288 (45.5)	1.7
3B	CB3	40	3,964 (140)	3.3
4B	CB4	15	1,487 (52.5)	6.0
5B	CB5	20	1,982 (70)	21.1
6B	CB6	15	1,487 (52.5)	0.7

Table 4-2 Cleanup Air Samples

Air Sample ID	Pellet ID	Sample Run Time (min)	Total Volume Liters (ft³)	Alpha Activity (dpm)
1C	CB1	20	1,982 (70)	7.7
2C	CB2	20	1,982 (70)	16.7
3C	CB3	10	991 (35)	4.0
4C	Control	-	-	-
5C	CB5	10	991 (35)	1.0
6C	CB6	10	991 (35)	0.7

*There was no cleanup air sample for pellet CB4 due to application of the class D extinguishing media.

The sample run times in Tables 4-1 and 4-2 provide the time lapse data for each evolution (oxidation and cleanup) in the separate trials.

4.2 Analytical Measurements

Following the air sample analysis protocols, the filters were sent to GEL Laboratories LLC for analytical measurement via gamma and alpha spectroscopy. Gamma spectroscopy results confirmed no detectable activity. A positive activity indication or radionuclide identification would have indicated a contaminant in the experiment. Air sample 4C corresponding to what would have been the cleanup air sample for pellet CB4 was submitted as a control air sample to establish a numerical background value. The laboratory reported the quantity of radioactivity and the total combined error as appropriate even if designating the result as “non-detectable.” This practice helps to avoid introducing a negative conceptual bias into the information ultimately reported.

The air samples were analyzed via alpha spectroscopy for ^{233/234}U, ^{235/236}U and ²³⁸U; these results are summarized in Tables 4-3 and 4-4.

Table 4-3 Analytical Laboratory. Oxidation Air Sample Alpha Spec. Data

Air Sample	Pellet ID	Analysis Type (α-spec)	Sample Value (dpm/sample)	TPU 1-Sigma (dpm/sample)	MDA (dpm/sample)
1B	CB1	Uranium	0.614	0.0767	0.106
		233/234	0.102	0.0381	0.0936
		Uranium	0.233	0.0425	0.0205
		235/236			
		Uranium 238			
2B	CB2	Uranium	0.271	0.0557	0.105
		233/234	0.0783	0.0368	0.0938
		Uranium	0.879	0.0957	0.0759
		235/236			
		Uranium 238			
3B	CB3	Uranium	0.722	0.0912	0.103
		233/234	0.203	0.0499	0.032
		Uranium	2.87	0.213	0.0955
		235/236			
		Uranium 238			
4B	CB4	Uranium	1.6	0.137	0.0999
		233/234	0.157	0.0449	0.0884
		Uranium	3.04	0.211	0.0826
		235/236			
		Uranium 238			
5B	CB5	Uranium	3.64	0.252	0.11
		233/234	0.529	0.0825	0.0994
		Uranium	19.3	1.03	0.0642
		235/236			
		Uranium 238			
6B	CB6	Uranium	0.333	0.0619	0.109
		233/234	0.142	0.0437	0.0779
		Uranium	1.05	0.107	0.0247
		235/236			
		Uranium 238			

Table 4-4 Analytical Laboratory. Cleanup Air Sample Alpha Spec. Data

Air Sample	Pellet ID	Analysis Type (α-spec)	Sample Value (dpm/sample)	TPU 1-Sigma (dpm/sample)	MDA (dpm/sample)
1C	CB1	Uranium	2.63	0.222	0.159
		233/234	0.267	0.0669	0.102
		Uranium	6.01	0.405	0.156
		235/236 Uranium 238			
2C	CB2	Uranium	3.29	0.257	0.137
		233/234	0.465	0.0843	0.0399
		Uranium	18.6	1.06	0.119
		235/236 Uranium 238			
3C	CB3	Uranium	0.361	0.0657	0.141
		233/234	0.193	0.0489	0.097
		Uranium	2.43	0.176	0.0873
		235/236 Uranium 238			
4C	Control	Uranium	0.0529	0.0332	0.0995
		233/234	0.0397	0.0397	0.0976
		Uranium	0.0234	0.0234	0.0631
		235/236 Uranium 238			
5C	CB5	Uranium	0.792	0.11	0.188
		233/234	0.149	0.0559	0.137
		Uranium	1.11	0.122	0.096
		235/236 Uranium 238			
6C	CB6	Uranium	0.745	0.101	0.189
		233/234	0.212	0.0515	0.0772
		Uranium	0.979	0.103	0.0782
		235/236 Uranium 238			

4.3 Analysis and Hypothesis Testing

Trial one was a proof of ignition test. Trial two and three were the cases in which secondary ignition of bulk DU metal was tested with the metal being placed below the pellet for Trial two and on top of the pellet for Trial three. Trail four and five investigated different application methods of class D extinguishing media. Trial six was the high-density pellet.

4.3.1 Analysis

Prior to determining the ARFs a comparative analysis between the alpha activity reported by the field measurements was made to the alpha spectroscopy results from the analytical laboratory as seen in Tables 4-5 and 4-6. The percent difference was calculated in the following way:

$$\% \text{ Difference} = \frac{(\text{Field Sample} - \text{Analytical Sample})}{\text{Analytical Sample}} \times 100 \quad \text{eq. (3)}$$

Table 4-5 Air Sample Activity Comparison for the Oxidation Trials

Air Sample	Pellet ID	Field Sample Value (dpm/filter)	Analytical Sample Value (dpm/filter)	Fraction (Field / Analytical)	% Difference
1B	CB1	1.0	0.95	1.05	5%
2B	CB2	1.7	1.23	1.38	38%
3B	CB3	3.3	3.80	0.87	-13%
4B	CB4	6.0	4.80	1.25	25%
5B	CB5	21.1	23.47	0.90	-10%
6B	CB6	0.7	1.53	0.46	-54%

Table 4-6 Air Sample Activity Comparison for the Cleanup

Air Sample	Pellet ID	Field Sample Value (dpm/filter)	Analytical Sample Value (dpm/filter)	Fraction (Field / Analytical)	% Difference
1C	CB1	7.7	8.91	0.86	-14%
2C	CB2	16.7	22.36	0.75	-25%
3C	CB3	4.0	2.98	1.34	34%
4C	Control	-	0.178	-	-
5C	CB5	1.0	2.05	0.49	-51%
6C	CB6	0.7	1.94	0.36	-64%

Large percent differences and fractions between the field sample value and the analytical sample values are expected when there is little activity available to measure. It is a simple comparison to make but yielded little useful information for this case. Comparison of the mean activities between the two measurement methods provides more informative and shows a range of agreement in the population means of 6% and 21% as illustrated in Table 4-7.

Table 4-7 Mean Alpha Activity Comparison (Field to Analytical Measurement)

Trial	Field Mean (dpm/sample)	Analytical Mean (dpm/sample)	% Difference in Means (Field to Analytical)
Oxidation Trials	5.63	5.86	6%
Cleanup Trials	6.02	7.65	21%

It is expected that had there been higher activity samples, the percent differences would be quite small between the two measurement results. The field counting equipment ensures compliance with the INL Radiological Controls Program but does not have the sensitivity for determination of ARFs from low activity air samples. Therefore, the remaining analysis and ARF determinations are based entirely on the alpha spectroscopy results.

Prior to determining the ARFs the activity reported for each alpha spectroscopy results were converted to mass equivalency by eq. (4).

$$Total\ Uranium\ Mass\ (g/sample) = \sum_{i=1}^n \left(\frac{Alpha\ Spec\ Result\ (dpm/sample)_i}{2.22 \times 10^{12} dpm/Ci} \right) \frac{1}{Specific\ Activity\ (Ci/g)_i} \quad eq. (4)$$

where,

$i = {}^{234}U, {}^{235}U\ and\ {}^{238}U$

Equation four was also used in converting the 1-sigma combined standard uncertainty units reported by the analytical laboratory for each alpha spectroscopy result. Then the individual combined standard uncertainties were propagated by the square root of the sum of the squares technique per eq. 5.

$$Total\ Uranium\ Uncertainty\ (g/sample) = \sqrt{(\sigma_{234}^2 + \sigma_{235}^2 + \sigma_{238}^2)} \quad \text{eq. (5)}$$

where

$\sigma_x^2 = \text{one} - \text{sigma combined standard uncertainty reported in grams per sample}$

The first ARF calculated is for the oxidation reactions. The oxidation reactions were defined as being from the time shortly before the removal of the crucible lid until the cessation of the reaction as determined by the absences of heat after complete mechanical agitation of the resultant oxidized pellet. The ARF for the oxidation reaction and its uncertainty are determined by dividing the result of eq. (4) and eq. (5) for each trial by the pellet mass, respectively. See eq. (6) and the results illustrated in Figure 4-1, error bars at one-sigma combined total uncertainty is included on all data points in the figure, but most are not visible given their small numerical value and the log-scale presentation.

$$ARF_{ox} = \frac{\sum_{i=1}^n \left(\frac{Alpha\ Spec\ Result\ (dpm)_i}{2.22 \times 10^{12} \text{ dpm/Ci}} \right)}{Total\ Pellet\ Mass\ (g)} \pm \frac{\sqrt{(\sigma_{234}^2 + \sigma_{235}^2 + \sigma_{238}^2)}}{Total\ Pellet\ Mass\ (g)} \quad \text{eq. (6)}$$

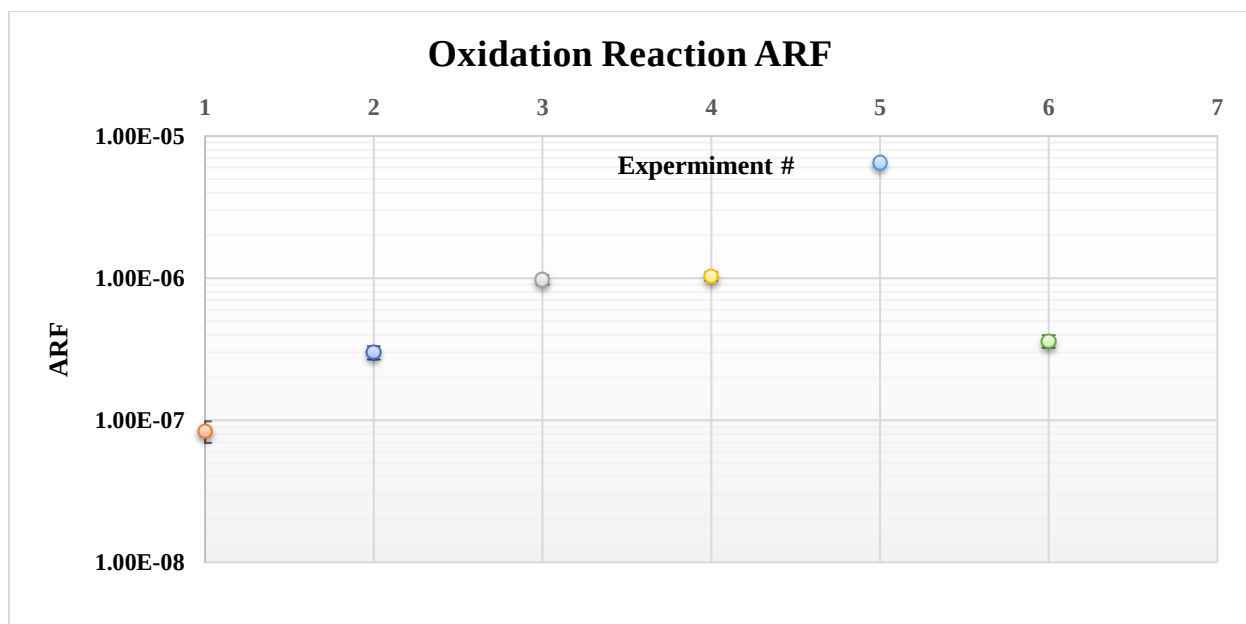


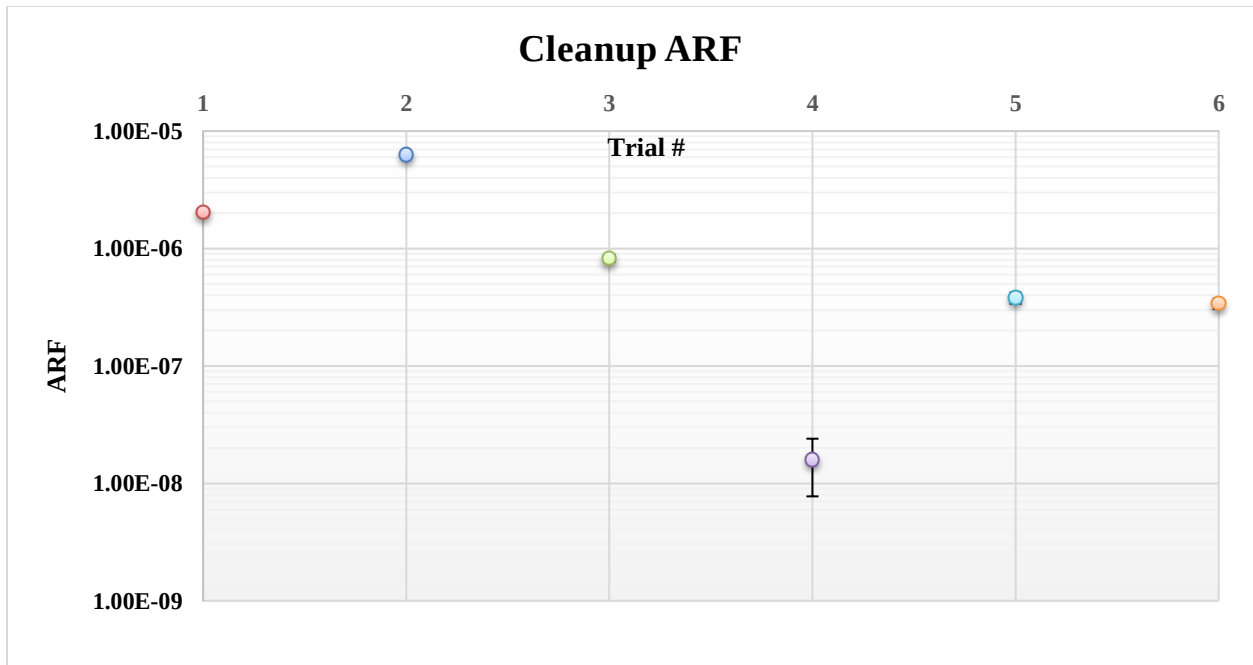
Figure 4-1 Oxidation ARFs

Next the ARF for the cleanup was determined. The cleanup was defined as the time-period beginning after the completion of the oxidation reaction and ending after the reaction stage and all surrounding areas had been returned to radiological clean levels (i.e., no detectable alpha or beta contamination).

Although not necessary for such small masses, in striving for completeness, a modification to the calculation of the ARF for the cleanup was made to account for the mass lost during the oxidation reaction per eq. (7). The results are illustrated in Figure 4-2, error bars at one-sigma combined total uncertainty are included on all data points in the figure, but most are not visible given their small numerical value and the log-scale presentation.

$$ARF_{cu} = \frac{\sum_{i=1}^n \left(\frac{\text{Alpha Spec Result (dpm)}_i}{2.22 \times 10^{12} \text{ dpm/Ci}} \right)}{\text{Specific Activity (Ci/g)}_i} \quad \text{eq. (7)}$$

$$\pm \frac{\sqrt{(\sigma_{234}^2 + \sigma_{235}^2 + \sigma_{238}^2)}}{[\text{Total Pellet Mass} - \text{Filter Mass Oxidation}]}$$



Note: Trial #4 is the control air filter. It is included for completeness and demonstrates the change in uncertainty when no activity is present.

Figure 4-2 Cleanup ARFs

Figure 4-3 plots both evolutions for each trial for comparison of the ARFs. Error bars at one-sigma combined total uncertainty are included on all data points in the figure, but most are not visible given their small numerical value and the log-scale presentation.

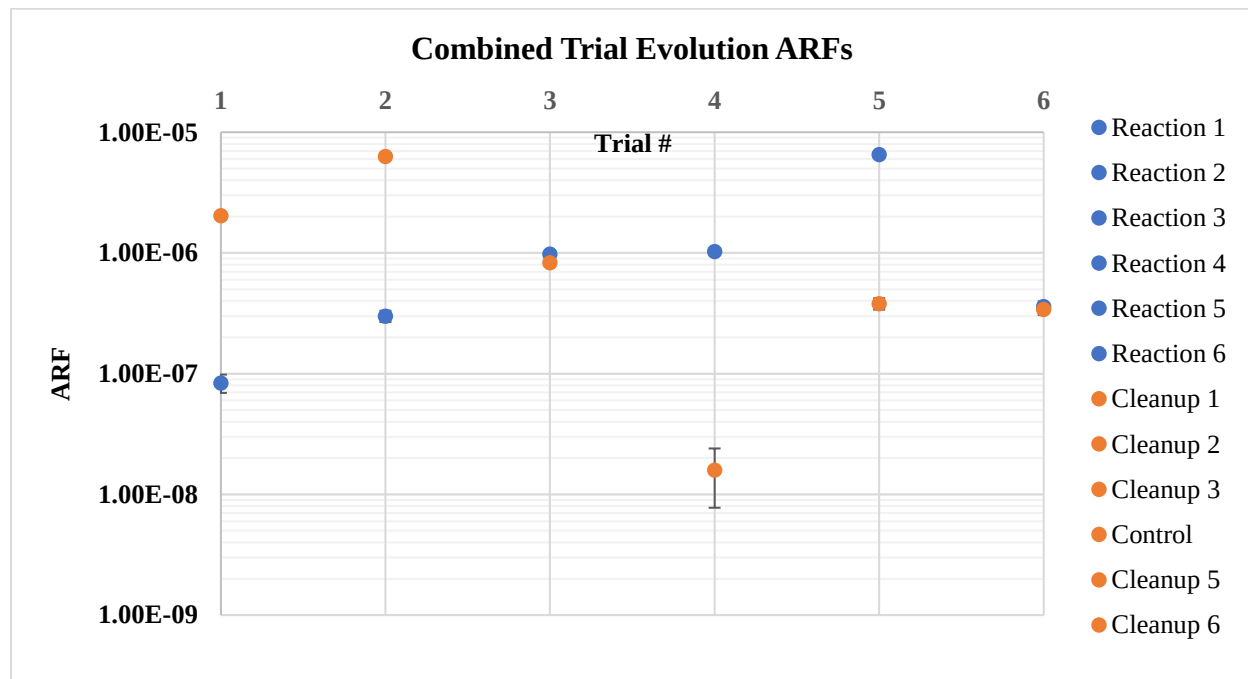


Figure 4-3 Combined Trial Evolution ARFs

Figure 4-4 is the total ARF for each trail (oxidation + cleanup) as calculated by eq. (8).

$$Total\ ARF = ARF_{ox} + ARF_{cu} \pm \sqrt{\sigma_{ARF_{ox}}^2 + \sigma_{ARF_{cu}}^2} \quad \text{eq. (8)}$$

Total uncertainty was calculated for the results in Figure 4-4 and are listed in Table 4-8 as the combined total uncertainty was too small to be visible as error bars in the figure.

Table 4-8 Total ARF and Total ARF 1-Sigma

Trial #	Pellet ID	Total ARF	Total ARF 1-sigma
1	CB1	2.12x10 ⁻⁰⁶	1.37x10 ⁻⁰⁷
2	CB2	6.58x10 ⁻⁰⁶	3.58x10 ⁻⁰⁷
3	CB3	1.80x10 ⁻⁰⁶	9.30x10 ⁻⁰⁸
4	CB4	1.03x10 ⁻⁰⁶	7.09x10 ⁻⁰⁸
5	CB5	6.89x10 ⁻⁰⁶	3.48x10 ⁻⁰⁷
6	CB6	7.01x10 ⁻⁰⁷	5.01x10 ⁻⁰⁸

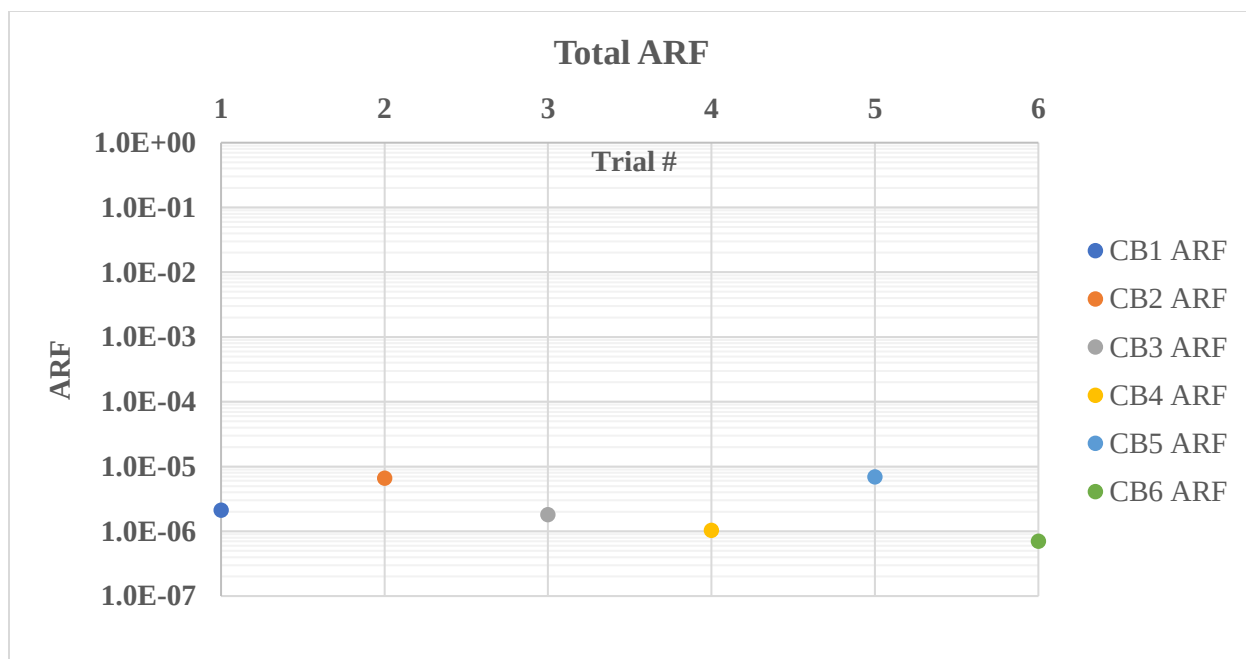


Figure 4-4 Total ARFs (Oxidation + Cleanup) per Trial

For each trial, a calculated ARF is considered to have a statistical difference from the other calculated ARF value when the calculated values reside outside the one sigma total combined uncertainties. Explicitly stated, a discernable difference exists when there is no overlap in the one sigma total combined uncertainty of the two calculated ARF values. Table 4-9 summarizes the statistical (discernable) differences in the calculated ARF values between trial processes (oxidation or cleanup).

Table 4-9 Statistical (Discernable) Difference in ARFs

Process	Trial	ARF	ARF - 1σ	ARF + 1σ	Discernable Difference
<i>Oxidation</i>	1	8.37x10 ⁻⁸	6.93x10 ⁻⁸	9.82x10 ⁻⁸	Yes
<i>Cleanup</i>	1	2.04x10 ⁻⁶	1.90x10 ⁻⁶	2.17x10 ⁻⁶	
<i>Oxidation</i>	2	3.00x10 ⁻⁷	2.67x10 ⁻⁷	3.32x10 ⁻⁷	Yes
<i>Cleanup</i>	2	6.28x10 ⁻⁶	5.92x10 ⁻⁶	6.64x10 ⁻⁶	
<i>Oxidation</i>	3	9.76x10 ⁻⁷	9.04x10 ⁻⁷	1.05x10 ⁻⁶	Yes
<i>Cleanup</i>	3	8.27x10 ⁻⁷	7.68x10 ⁻⁷	8.87x10 ⁻⁷	
<i>Oxidation</i>	4	1.03x10 ⁻⁶	9.59x10 ⁻⁷	1.10x10 ⁻⁶	Yes
<i>Cleanup</i>	Control	1.59x10 ⁻⁸	7.75x10 ⁻⁹	2.40x10 ⁻⁸	
<i>Oxidation</i>	5	6.51x10 ⁻⁶	6.16x10 ⁻⁶	6.86x10 ⁻⁶	Yes
<i>Cleanup</i>	5	3.81x10 ⁻⁷	3.40x10 ⁻⁷	3.81x10 ⁻⁷	
<i>Oxidation</i>	6	3.60x10 ⁻⁷	3.24x10 ⁻⁷	3.96x10 ⁻⁷	No
<i>Cleanup</i>	6	3.40x10 ⁻⁷	3.06x10 ⁻⁷	3.75x10 ⁻⁷	

The “discernable difference” column in Table 4-9 is determined by direct comparison of the calculated ARF value and the associated one-sigma uncertainty. The calculated ARF values are compared to determine which is the smaller ARF value. Once the smaller value is determined then the upper limit of the uncertainty is compared to the lower limit of the uncertainty associated with the larger ARF value. If the upper limit uncertainty has a larger numerical value than the lower limit uncertainty, then the uncertainty of the two calculated ARF values overlap at one-sigma. An overlap of the uncertainties indicates that the values are statistically the same number. This is illustrated in the trial six outcome.

4.3.2 Hypothesis Testing

The following hypotheses were tested in this research the numerical quantities of the P-value statistic for hypothesis testing. The P-value is the probability of getting a sample statistic (i.e., the mean) or a more extreme sample statistic in the direction of the alternative hypothesis when the null hypothesis is true (Bluman). The α level (value) of 0.05 was chosen for the probability of getting a type I error and the t Test was utilized given $n < 30$.

$$t = \frac{\bar{X} - \mu}{s / \sqrt{n}} \quad \text{eq. (9)}$$

where:

$\bar{X}$ = sample mean

μ = population mean

s = sample standard deviation

n = sample size

The p-value was calculated using a TI-83 plus calculator with associated statistics solver instead of the traditional look and interpolation method utilizing the t -statistic and α -value. The decision rule when using a p-value is given as:

If p-value $\leq \alpha$, reject the null hypothesis.

If p-value $> \alpha$, do not reject the null hypothesis (Bluman).

Test 1: Depleted uranium (DU) oxidation process without extinguishing media applied:

$H_{1,0}$: There is not a difference in the ARFs between the oxidation process and the cleanup process ($H_{1,0}$: $\mu_{ox} - \mu_{cu} = 0$).

$H_{1,A}$: There is a difference in ARFs between the oxidation process and the cleanup process ($H_{1,A}$: $\mu_{ox} - \mu_{cu} \neq 0$).

For Test-1, the parameters set to evaluate the data were assessed with a paired sample t-test at a statistical cutoff $\alpha = 0.05$. Given the p-value (0.268) is greater than the statistical cutoff α , the data is consistent with the null hypothesis. See Table 4-10.

Table 4-10 Test 1: Hypothesis Testing DU Oxidation without Extinguishing Media Applied

Trial	ARF_{ox}	ARF_{cu}	ARF_{ox} - ARF_{cu}
1	8.37x10 ⁻⁸	2.04x10 ⁻⁶	-1.95x10 ⁻⁶
2	3.00x10 ⁻⁷	6.28x10 ⁻⁶	-5.98x10 ⁻⁶
3	9.76x10 ⁻⁷	8.27x10 ⁻⁷	1.48x10 ⁻⁷
6	3.6x10 ⁻⁷	3.40x10 ⁻⁷	2.02x10 ⁻⁸
Mean of the Differences			-1.94x10 ⁻⁶
Standard Deviation of the Differences			2.86x10 ⁻⁶
T-statistic			-1.36
P-Value			0.268

Test 2: DU oxidation process with extinguishing media applied:

$H_{2,0}$: There is not a difference in the ARFs between the oxidation process and the cleanup process

$$(H_{2,0}: \mu_{ox} - \mu_{cu} = 0).$$

$H_{2,A}$: The highest ARFs occurs during the oxidation process ($H_{2,A}: \mu_{ox} - \mu_{cu} > 0$).

For Test-2, the parameters set to evaluate the data were assessed with a paired sample t-test at a statistical cutoff $\alpha = 0.05$. Given the p-value (0.197) is greater than the statistical cutoff α , the data is consistent with the null hypothesis. See Table 4-11.

Table 4-11 Test 2: Hypothesis Testing DU Oxidation with Extinguishing Media Applied

Trial	ARF_{ox}	ARF_{cu}	ARF_{ox} - ARF_{cu}
4	1.03x10 ⁻⁶	0	1.03x10 ⁻⁶
5	6.51x10 ⁻⁶	3.81x10 ⁻⁷	6.13x10 ⁻⁶
Mean of the Differences			3.58x10 ⁻⁶
Standard Deviation of the Differences			3.61x10 ⁻⁶
T-statistic			1.40
P-Value			0.197

Test 3: Application of the extinguishing media affects the ARFs:

$H_{3,0}$: The application of extinguishing media does not affect the total ARF

$$(H_{3,0}: \mu_{w/o\ ext} - \mu_{w-ext} = 0).$$

$H_{3,1}$: The application of extinguishing media does affect the total ARF

$$(H_{3,1}: \mu_{w/o\ ext} - \mu_{w-ext} \neq 0).$$

For Test-3, the parameters set to evaluate the data were assessed with a two-sample t-test at a statistical cutoff $\alpha = 0.05$. Given the p-value (0.76) is greater than the statistical cutoff α , the data is consistent with the null hypothesis. See Table 4-12.

Table 4-12 Test 3: Hypothesis Testing Application of Extinguishing Media Affects the Total ARF

Without Extinguishing Media		With Extinguishing Media	
<i>Trial</i>	<i>Total ARF</i>	<i>Trial</i>	<i>Total ARF</i>
1	2.12x10 ⁻⁶		
2	6.58x10 ⁻⁶	4	1.03x10 ⁻⁶
3	1.80x10 ⁻⁶	5	6.89x10 ⁻⁶
6	7.01x10 ⁻⁷		
Sample Mean	2.80x10 ⁻⁶	Sample Mean	3.96x10 ⁻⁶
Sample Standard Deviation	2.59x10 ⁻⁶	Sample Standard Deviation	4.14x10 ⁻⁶
T-statistic = -0.36			
P-Value = 0.764			

There was no cleanup ARF of the fourth trial due to the rapid extinguishing of the oxidation reaction by application of the class D extinguishing media. The application method used a plastic bag with extinguishing media inside placed over the oxidizing pellet. This resulted in the immediate cessation of the reaction by encapsulation of the pellet in the extinguishing media. The product of this approach was no spread of contamination. The data represented in the figures and tables for cleanup of trial four is the control alpha spectroscopy air-sample data, as reported by the analytical laboratory. For hypotheses testing purposes, the value for the ARF in Trial-four was set to zero for completeness. Albeit there is no

significant change in the hypotheses test parameters when the reported activity is used instead of the zero value.

For trial five the class D extinguishing media was liberally applied by shaker can and followed by significant mechanical agitation. This process was repeated multiple times prior to initiating the cleanup process.

5 Summary and Conclusion

5.1 Summary

The estimates of the ARFs derived in this work are under a specific set of conditions that are understood to have large unquantifiable uncertainties. For instance, the air sampler may have been started minutes (up to 2 minutes) prior to the actual oxidation event thus causing a dilution of the concentration. This is one reason the total mass accumulated on the filters was utilized and not the concentration as would be typical in most air monitoring situations when a minimum known activity concentration is being determined for purposes of personnel and facility protection.

Another source of uncertainty is that the experimental setup is an open system, whereas most studies of airborne release fractions utilize closed systems to ensure no unnecessary loss (dilution) of airborne material. While a closed system was not utilized in this research, the conditions of sampling are believed by the investigators to more closely reflect those of most operational air monitoring situations and therefore the information derived is of importance.

This research project, being purely an Operational Health Physics project, has determined ARFs (approximately 10^{-6}) consistent with benchmark historical studies (utilizing closed systems) that were specifically undertaken for determination of ARF values.

The goal of this research was to better understand potential ARF values associated with advanced fuel fabrication research conducted at the INL. ARF values were investigated for oxidation and cleanup including when different application methods of class D extinguishing media were utilized. ARF values were determined for each individual trial process evolution (oxidation and cleanup) as well as a total ARF determination for each trial. Correlation of the individual data points for trial and trial evolutions between field measurements and analytical measurements proved difficult and inconclusive based on the low levels of measurable activity. Comparison of the mean activities between the two measurement methods

proved more informative with a mean population difference in the trials of 6% for oxidation process and 21% for the cleanup process.

Hypotheses tests were performed but given such a limited data set there was not enough statistical evidence to reject any of the stated null hypotheses.

In this research ARF values for DU never exceeded 1×10^{-5} for any single trial evolution and total ARFs fluctuated between and maximum of 7×10^{-6} to a minimum of 7×10^{-7} . The ARF values determined in this research effort agree well with early similar studies and are well below the bounding ARF value 1×10^{-3} and median value 1×10^{-4} prescribed in DOE-HDBK-3010-94.

5.2 Future Work

Given the operational nature of this research project, the data set was limited and there was never an opportunity to perform particle size distribution analysis. For future work it is recommended that a larger number of trials be performed to give more statistical power when performing hypothesis testing. Additionally, it is recommended that an apparatus for particle size determination be designed and employed so that an assessment of the respirable fraction be made for use in conjunction with the ARFs determined here.

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