DEFENSE NUCLEAR FACILITIES SAFETY BOARD

Washington, DC 20004-2901

September 7, 2018

The Honorable James Richard Perry
Secretary of Energy
U.S. Department of Energy
1000 Independence Avenue, SW
Washington, DC 20585-0701

Dear Secretary Perry:

The Defense Nuclear Facilities Safety Board's staff has reviewed the experimental results of a Nuclear Safety Research and Development project at the Y-12 National Security Complex. The experimental project is designed to determine the uranium airborne release fraction (ARF) and respirable fraction (RF) to be used in safety analysis of fires at Y-12 defense nuclear facilities. Our staff identified technical concerns with this project and the use of its preliminary results in the Uranium Processing Facility (UPF) safety analysis. These concerns indicate that use of ARF and RF values derived from the Y-12 experiments would likely lead to underestimating the dose consequences of accidents involving uranium metal fires which could result in the selection of inadequate safety controls. These concerns are discussed in the enclosed report.

The enclosed analysis by our staff is provided for your information and use in evaluating the applicability of these results and in considering revisions to the DOE Handbook 3010-94, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities*.

Yours truly,

Bruce Hamilton
Acting Chairman

Enclosure

c: Mr. Joe Olencz

DEFENSE NUCLEAR FACILITIES SAFETY BOARD

Staff Issue Report

July 3, 2018

MEMORANDUM FOR: C. J. Roscetti, Technical Director

COPIES: Board Members

FROM: J. Abrefah

SUBJECT: Airborne Release Parameters for Uranium Alloys

Members of the Defense Nuclear Facilities Safety Board's (Board) technical staff reviewed the results of a research project at the Y-12 National Security Complex (Y-12) to determine airborne release fraction (ARF) and respirable fraction (RF) values for metallic uranium and its alloys involved in fires. Y-12 performed the research as part of the Nuclear Safety Research and Development (NSR&D) program [1–4]. The focus of the experimental work was to determine ARF and RF values for Y-12's inventory of metallic uranium that could be used in the safety analysis of potential fires in a defense nuclear facility. Y-12 expected its experimental results to justify using lower values of ARF*RF for its safety analysis instead of the bounding value of 1E-3 listed in Department of Energy (DOE) Handbook DOE-HDBK-3010-94, (Handbook 3010), *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities* [5].

Background. DOE safety analysts use ARFs and RFs (or their product, ARF*RF) in calculating the dose consequences of radionuclides released in accidents involving a nuclear facility, such as fires. The DOE guidance for choosing ARF and RF values for various accident conditions and radionuclide material forms is found in Handbook 3010. Handbook 3010 was principally authored by Jofu Mishima and D. Pinkston of the Science Application International Corporation, who used experimental results available at the time to derive bounding values for ARF and RF. For uranium metal under thermal stress, the bounding release fractions in Handbook 3010 were derived from experiments conducted by Elder and Tinkle [6] at Los Alamos National Laboratory on samples of a beta-phase stabilized alloy of depleted uranium with 0.75 weight-percent (wt%) titanium. The recommended bounding ARF*RF in Handbook 3010 for uranium metals is 1E-3.

Elder and Tinkle Tests—The Elder and Tinkle tests investigated the nature and amount of uranium oxide particulates that become airborne if depleted uranium penetrators (anti-tank munitions, designated XM774) were subjected to fire in a storage depot or during transport. In their experiments, Elder and Tinkle exposed the uranium penetrators to high temperatures and an oxidizing atmosphere, with a wind speed of 2.23 m/s (5 mph). Elder and Tinkle performed laboratory experiments using an electrically heated furnace, as well as outdoor experiments where the uranium penetrators were placed in proximity to flames from burning materials. Elder and Tinkle estimated that approximately six to thirty percent of the uranium was oxidized in their laboratory experiments, over the course of two to four hours. Their outdoor experimental burn test #4, which was the basis for the bounding ARF*RF in Handbook 3010, resulted in 42–47 weight percent of the uranium penetrators oxidized over the course of about three hours. The fuel in that test consisted of pine wood and packing tube material, added in ten batches for the burn time of three hours.

The Elder and Tinkle experiments, as well as other experiments [7–11] provide insights into the process for generating uranium oxide aerosols from uranium metals during a fire event. As the uranium metal is exposed to high temperatures from the fire, different species of uranium oxide form on the surface of the uranium metal sample. The extent of oxidation is influenced by factors including temperature, exposure time, and the uranium sample's surface area. Some of the uranium oxide will spall off from the uranium sample, forming particulates. A small fraction of the generated particulates will become airborne. Spallation is related to stresses, such as those resulting from the change in density that occurs at the reaction interface as metal is converted to oxide. Also, changes in temperature can cause thermal stresses in the oxide layer, so temperature cycling can lead to increased spallation and aerosol generation.

Elder and Tinkle's principal conclusion from their tests was that uranium "aerosols with particles in the respirable size range are produced when the uranium penetrators were exposed to temperatures above 500°C for times on the order of one-half hour or more." The authors concluded that the production of the uranium oxide and its aerosol is enhanced by forced draft and temperature cycling during exposure of uranium under fire conditions.

*DOE Handbook 3010 ARF*RF*—While the uranium samples in the Elder and Tinkle tests did not fully oxidize, the authors of Handbook 3010 chose to extrapolate the measured results to derive a bounding value of ARF*RF that is applicable to a fully oxidized mass of uranium. The authors used the highest concentration of uranium oxide aerosol observed during experimental outdoor burn test #4 to perform this extrapolation. The result was the bounding ARF*RF value of 1E-3 for bulk uranium metal under thermal stress.

Y-12 NSR&D ARF and RF Experiments. The Engineering and Applied Technologies Division of the Y-12 management and operating contractor (at that time, BWXT, LLC) performed experiments to derive ARF and RF values for Y-12's inventory of metallic uranium, including alloys [1–4]. BWXT personnel designed an experimental system to characterize the mass and size distribution of uranium oxide particles that would be airborne when uranium metal alloys were oxidized during a fire event in a storage building and/or process facility. BWXT was motivated to perform these tests [1, 3] because of (a) limited data have been published on aerosol production from large pieces of uranium metal, (b) the physical forms and type of alloys used in the reference experiments of Handbook 3010 are not in use at Y-12, and (c) the response of Y-12 uranium alloy materials under experimental conditions similar to the referenced tests in Handbook 3010 may yield lower ARF and RF values compared to the handbook bounding values.

The experimental system design used a four-inch diameter schedule-40 stainless steel pipe to construct the burn chamber, with a quartz window for visual examination of the burning process. The BWXT personnel used three Kingsford® brand charcoal briquettes, with a total weight of about 70 g, as the combustible material to provide flame impingement to the 0.3–1 kg uranium samples. Thermocouples in contact with the top and bottom of the uranium sample monitored the temperature history during the test. The completed tests at the time of the staff team's review used samples of: (a) alpha-phase uranium metal, (b) beta-phase uranium-titanium alloy (0.75 wt% U), and (c) gamma-phase uranium-niobium alloy (6 wt% Nb).

BWXT personnel conducted five experimental tests for each type of uranium metal or alloy. The experimenters measured the initial mass of the uranium sample, as well as the post-fire mass of the sample after easily removed oxide was brushed off. They used the change in mass as an indication of how much of the metal was oxidized in each run. Air flow of approximately 14 standard-cubic-feet-per-minute through the bottom of the test assembly carried uranium oxide particles to a sampling chamber where a Tisch Eight-Stage Cascade Impactor with a sampling nozzle was used to determine the RF. The bulk of the uranium oxide particles that remained airborne were collected by a gross filter sampler. The experimenters measured the mass of uranium oxides captured in the sampling chamber, including the cascade impactor and the gross filter. They calculated the ARF for each run as the uranium mass for the uranium oxide recovered in the sampling chamber, divided by the initial uranium mass of the sample. The results are available in reports and publications authored by BWXT and Babcock & Wilcox Technical Services Y-12, LLC personnel [3, 4].

The change in the uranium sample mass was relatively small, indicating that very little uranium was oxidized. The loss in mass resulting from spallation of uranium oxide from the alpha uranium, for example, ranges from 0 to less than 0.1 weight percent. For the tests involving the uranium-niobium alloy, the sample sometimes showed a small mass gain due to the adherent oxide product. Due to the oxide layer that remained on the samples and the low amount of spallation, it is difficult to accurately determine quantitative values for the extent of oxidation. The measured mass of airborne uranium oxide product in the tests was also very small; only tens of micrograms were collected for the uranium-niobium tests.

The Y-12 experimenters reported the mean ARF*RF for the different metals tested, based on the five runs for each metal [4]. The Consolidated Nuclear Security, LLC (CNS) safety analysts supporting the Uranium Processing Facility (UPF) project rounded those results to obtain the following ARF*RF values for the safety analysis [12] of postulated UPF fire events:

- Alpha-phase uranium, 1E-6
- Uranium-titanium alloy, 1E-5
- Uranium-niobium alloy, 1E-7

Board's Staff Evaluation of Y-12 Experimental Results. The Board's staff review team agrees that improving the technical basis of ARF and RF values used in safety analysis is an appropriate focus for nuclear safety research and development. The scientific literature provides some support to the BWXT hypothesis that different alloys of uranium may exhibit different oxidation behavior at high temperatures [13-15]. Thus there is value to performing

ARF and RF experiments on different alloys of uranium. Further, the bounding Handbook 3010 values for uranium under thermal stress are based on limited data, so additional experiments could be beneficial.

The staff team reviewed the Y-12 experimental program and the resulting ARF*RF values. The staff team evaluated whether the ARF*RF values are applicable to the safety analysis of bounding design basis fires. The staff team also evaluated whether the test program and apparatus produced consistent and representative results.

Comparison to Bounding Handbook 3010 Value—Given that the bounding ARF*RF in Handbook 3010 was determined from the Elder and Tinkle tests involving a uranium-titanium alloy, a major purpose of the Y-12 tests was to determine how other uranium alloys might behave differently. The Y-12 program also included the uranium-titanium alloy, which provided the staff team with a basis for comparing the Y-12 results and analysis to those in Handbook 3010. The staff team found that the Y-12 program did not perform testing comparable to that of Elder and Tinkle, or analysis comparable to that in Handbook 3010. In the key Elder and Tinkle test, pieces of uranium-titanium alloy (about 3.3 kg each) were 42-47% oxidized. The authors of Handbook 3010 then extrapolated the Elder and Tinkle results to derive an ARF*RF value, 1E-3, that corresponds to a fully oxidized piece of metal. By contrast, the Y-12 experimenters observed much less oxidation in their uranium-titanium samples. The experimenters estimated that their samples (which were smaller, at about 0.3 to 1 kg each) were only 0.1 to 0.4% oxidized. As noted above, CNS safety analysts have derived an ARF*RF of 1E-5 that reflects the low level of oxidation observed in the tests at Y-12 [12].

Thus, the ARF*RF value derived by Y-12 for the uranium-titanium alloy is much lower than what DOE considered to be a bounding value for that material in Handbook 3010. This observation leads the staff team to doubt whether the Y-12 apparatus and analysis led to ARF*RF values that can be considered bounding for use in safety analysis. The following sections provide additional detail.

Less Severe Fire Exposure in Y-12 Testing—The severity of the fire exposure differed between the Y-12 experiments and those of Elder and Tinkle, which is a likely reason for why the two sets of experiments had such different results for uranium-titanium. In the Elder and Tinkle test used for Handbook 3010, temperatures measured at the sample surface generally cycled between 700 and 900^{o}C over three hours, with peaks above 1100^{o}C [6]. In the Y-12 tests, the maximum temperature measured at the sample surface during any test run was about 415^{o}C. For many test runs, the peak temperature was below 350^{o}C, and for some test runs it was below 200^{o}C. The test runs lasted about an hour, compared to the three hours in Elder and Tinkle. Additionally, the tests performed by Elder and Tinkle had a great degree of temperature cycling due to the fuel being added in 10 batches. The Y-12 experiment did not have temperature cycling on this scale. The Y-12 tests led to ARF*RF values for uranium-titanium that are a factor of 100 lower than the Handbook 3010 value for the same alloy. The staff team concludes that the Y-12 experimenters did not design tests of the same robustness as the Elder and Tinkle tests.

Comparison to Design Basis Fires—While the Y-12 ARF*RF values are less conservative than the bounding value in Handbook 3010, they could still be appropriate for use if the severity of fire exposure in the test apparatus bounds the severity of Y-12 design basis fires. The staff team asked CNS experimenters how the test conditions compared to a design basis fire. The CNS experimenters compared the measured temperature history from the Y-12 experiments to the calculated temperature history for the Highly Enriched Uranium Materials Facility (HEUMF) design basis fire. The observed experimental temperatures bounded the temperatures postulated for the HEUMF design basis fire, so the CNS personnel concluded their experimental data should bound design basis fires at Y-12. Thus, the CNS personnel did not see the need to achieve more severe conditions in their tests, or to extrapolate from their results to more severe conditions.

The staff team has several observations regarding this reasoning:

- When CNS personnel compared their experimental temperatures to the HEUMF scenario, they chose the single bounding experimental run. Many of the other experimental runs would not have bounded the calculated HEUMF temperature data.

- The two data sets are not directly comparable. The calculated HEUMF data used by CNS personnel represented the upper gas layer in the room, which cannot be directly compared to the measured temperature at the test sample surface, which is near or within the flame in a test apparatus.

- The HEUMF fire scenario is relatively small compared to design basis fires at other DOE defense nuclear facilities, so the comparison performed by CNS personnel does not support the direct application of the Y-12 results elsewhere at Y-12 or across DOE.

- The Elder and Tinkle tests showed the importance of temperature cycling as a factor that should be considered for experiments or analysis of bounding scenarios. The Y-12 tests did not have the same temperature cycling.

The staff team concludes stronger technical basis is needed for comparing the Y-12 experimental results to the design basis fire of any specific nuclear facility. The technical basis should include clear criteria for determining whether the Y-12 experiment is applicable to the design basis fire of a given nuclear facility. Y-12 experimental data may be applicable to smaller, limited-scope fires, but without clear criteria for applicability, analysts could inappropriately apply the ARF*RF values derived by Y-12 to larger fires at other defense nuclear facilities at Y-12 or other DOE sites. This would lead to scenarios where accidents are not bounded.

A technically justified approach to extrapolate or extend the Y-12 results to a bounding scenario could be useful for safety basis development. The BWXT personnel showed [3] that when the Y-12 uranium-titanium ARF*RF values are extrapolated to full oxidation, they obtain values on the same order of magnitude as the bounding ARF*RF in Handbook 3010. This suggests that a method for predicting the extent of oxidation could be used in conjunction with the Y-12 results. Alternatively, the Y-12 results could be used to demonstrate relative differences in ARF*RF between the different alloys, but the actual values of ARF*RF could still be linked to the current value in Handbook 3010.

*Selection of ARF*RF Values*—The Y-12 experiments included five test runs for each metal or alloy. The ARF*RF varied between test runs for the same alloy. The BWXT experimenters calculated mean values of ARF*RF for each alloy, which CNS personnel rounded for use in UPF safety analysis. As a result, the ARF*RF values selected for UPF do not always bound the results measured in the Y-12 experiments. For example, CNS selected an ARF*RF of 1E-5 for the uranium-titanium alloy, which is notably smaller (less conservative/not bounding) than the ARF*RF of 6.6E-5 resulting from one of the uranium-titanium experimental runs.

Limitations of Y-12 Experiments—The BWXT experimenters explained to the staff team that they were aware of differences between their experimental apparatus and that of Elder and Tinkle. The Y-12 apparatus could not closely resemble Elder and Tinkle's due to environmental and safety requirements, as well as other practical constraints. The staff team understands that it may be difficult for the Y-12 experiments to achieve a more severe fire exposure. The constraints underscore the importance of identifying the limits of applicability of the Y-12 results.

Choice of Experimental Parameters—The pipe in the Y-12 experiments has different dimensions from that of Elder and Tinkle's apparatus. The BWXT researchers desired to set the flowrate of air through the apparatus in a way that would create conditions similar to that of Elder and Tinkle. They chose to scale the airflow by aiming to match the Reynolds number from the Elder and Tinkle experiment. The Reynolds number is useful for predicting whether flow will be laminar or turbulent. However, the BWXT experimenters calculated the Reynolds number for the Y-12 tests differently from the calculation they performed for the Elder and Tinkle test system. The Reynolds number includes a characteristic length for the flow. For the Elder and Tinkle experiments, they used the diameter of the uranium samples as the characteristic length. For the Y-12 experiments, they used the diameter of the pipe in which the air is flowing. In the former case, the Reynolds number is descriptive of the boundary layer that develops as the air flows over the uranium samples. In the latter case, the Reynolds number is descriptive of the internal flow of air in the pipe. As a result, the flow in the Y-12 experiments was likely in the laminar or transition regimes for most part of Y-12 piping outside the flame heated zone, whereas the flow in the Elder and Tinkle experiments would have been turbulent for the whole flow path of their apparatus. The staff team thus concludes that the method used by the Y-12 researchers to choose the air flowrate was not consistent for their system to reproduce the flow condition of the Elder and Tinkle apparatus.

Consistency of Results—The experimental results include some unexpected patterns. As described above, some of the airborne particles were collected in the cascade impactor, through a one-inch diameter nozzle that was located at the center of a four-inch diameter schedule-40 stainless steel pipe. The particles that were carried past that nozzle were captured on the gross filter. In most of the test runs, the majority of the airborne particles were recovered on the gross filter. However, in the uranium-niobium test runs and one of the alpha uranium test runs, most of the airborne particles were captured in the impactor. The BWXT experimenters did not investigate the cause of these anomalies. It is possible the anomalies were the result of a problem with the measurement, undetected differences in the airflow patterns, or some other unexpected phenomenon that could have affected the validity of the results. The staff team concludes these anomalies directly impacted the Y-12 data that was used to derive the experimental ARF and RF.

Oxidation Kinetics of Uranium and its Alloys—The BWXT experimenters used the varying oxidation resistance of different uranium metals and alloys to support performing tests to elucidate the potential thermal stress response for Y-12's inventory of uranium alloys.

In general, the chemistry of uranium and its alloys is complex, and accurate prediction of the uranium phase is complicated, due in part to the tendency of uranium to undergo multiple oxidation states. Environmental conditions such as temperature, humidity, and oxygen concentration have a strong influence on uranium materials, making the prediction of mechanistic and kinetic properties of oxidizing uranium and its alloys very difficult for an engineering test system. Similarly, the authors of Handbook 3010 noted that ARF experiments often do not completely characterize the full range of physical and chemical behavior that could be involved:

> *The generation and suspension of particles is the result of the interaction of multiple physiochemical variables that have not been completely characterized as the majority of the experiments performed were designed in an attempt to reflect reasonably bounding conditions for specific industrial situations of concern….Further, in many cases it is considered likely that accident specific ARFs are actually distributed in a highly irregular manner (i.e., multi-modal or truncated distributions).*

Considering these challenges, the staff team concludes that the Y-12 experiments cannot cover the full range of physical mechanisms and processes that could take place in actual facility fires. It is thus important to thoughtfully consider the limits of applicability of the Y-12 results. The authors of Handbook 3010 faced similar challenges of a limited uranium alloy data set in recommending bounding values for ARF and RF. This suggests that for a designed limited experimental program to generate ARF and RF data that have the technical basis to impact the bounding ARF*RF in the Handbook, the testing should adequately cover a variety of mechanistic experimental parametric conditions for extrapolation of the data to bounding conditions.

Incorporation of Y-12 Experimental Results in the UPF Safety Analysis. As mentioned above, UPF project personnel selected new ARF*RF values for the three types of uranium or uranium alloy tested in the Y-12 experiments. In practice, the UPF safety analysis documents reviewed by the staff team [12] only applied the Y-12 ARF*RF of 1E-6 for alpha-phase uranium. In the consequence analysis for the design basis fire event at UPF, much of the uranium in the facility was assigned to other material types, so the ARF*RF value of 1E-6 was only applied to a small subset of the total uranium inventory. Thus, using the derived Y-12 ARF*RF value instead of the Handbook 3010 value resulted in only a modest decrease in calculated dose consequences to collocated worker and off-site public. Because of the three orders of magnitude difference between the recommended Handbook 3010 ARF*RF value of 1E-3 and the derived Y-12 ARF*RF value of 1E-6, the use of the Y-12 ARF*RF values could affect safety-related decisions at other facilities, as well as at UPF in the future if changes occur to the inventory or analysis.

Conclusions. The staff team's review of the Y-12 NSR&D test results identified questions on whether the experimental program derived bounding ARF*RF values suitable for use in safety analyses for the UPF design and other DOE defense nuclear facilities. Because of these questions, the staff team concludes that using the current Y-12 NSR&D-derived ARF*RF values for a design basis fire event in the UPF safety analysis does not meet the intent of the DOE Standard 3009-94, *Preparation of Nonreactor Nuclear Facility Documented Safety Analysis*, which states that "calculations be based on reasonably conservative estimates of the various input parameters."

The staff review team notes that CNS plans to continue its NSR&D testing program to gather more data. These additional tests provide an opportunity to ensure that the derived ARF and RF values are bounding for safety analyses.

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