DEFENSE NUCLEAR FACILITIES SAFETY BOARD

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September 29, 1994

Mr. Mark Whitaker, EH-6
U.S. Department of Energy
1000 Independence Avenue, SW
Washington, D.C. 20585

Dear Mr. Whitaker:

Enclosed for your information and distribution are eight (8) Defense Nuclear Facilities Safety Board (DNFSB) staff reports. The reports have been placed in the DNFSB Public Reading Room.

Sincerely,

George W. Cunningham
Technical Director

Enclosures (8)

94-0005341

DEFENSE NUCLEAR FACILITIES SAFETY BOARD

February 22, 1994

MEMORANDUM FOR: G. W. Cunningham, Technical Director

COPIES: Board Members

FROM: Timothy J. Dwyer, Hanford Site Program Manager

SUBJECT: Report on Review of Plutonium Uranium Extraction (PUREX) Facility Material Unaccounted For (MUF)

1. **Purpose:** This report documents DNFSB staff observations from a review of DOE Richland Operations Office (DOE-RL) PUREX Facility MUF data conducted on site on November 2, and December 13, 1993, with subsequent document reviews and telephone conversations.

2. **Summary:** PUREX facility management has concluded that there is minimal hazard attributable to potential plutonium holdup in the plant as represented by calculated MUF values. The bases for this conclusion involve both the errors inherent in the MUF calculation itself, which were recognized to consistently overstate the amount of plutonium unaccounted-for, and the post-campaign flushes and inspections, which identified no appreciable build-up of plutonium occurring within the plant. Facility management stated that efforts to recover MUF during the 1990 cleanout run have minimized the potential for residual plutonium throughout PUREX. However, a review of PUREX MUF data demonstrates that since the final cleanout run conducted in 1990, kilogram quantities of plutonium have been "generated" yearly in PUREX. It will be necessary to understand the source of these kilogram quantities of plutonium and their implication with regard to worker safety during transition and decontamination and decommissioning (D&D) efforts.

3. **Background:** On August 31, 1993, members of the DNFSB Staff Transition Crosscutting Study Group participated in a video-teleconference with DOE headquarters, DOE-RL, and Westinghouse Hanford Company (WHC) representatives concerning the proposed transition activities scheduled to occur at the PUREX Facility. During the video-teleconference, a question was raised concerning the implications of past MUF occurrences on the proposed transition activities. Therefore, on November 2, 1993, Timothy J. Dwyer of the DNFSB Staff met with DOE-RL and WHC representatives at the PUREX facility to review available data. A follow-on meeting

was held on December 13, 1993, between Steven A. Stokes and WHC representatives to review photographic and video data.

4. **Discussion:** Material accountability at the PUREX Facility was based upon a deceptively simple mathematical formula:

$$\mathbf{MUF = input_{Rx\ code\ calculation} - output_{to\ PFP} - waste_{rough\ sample\ only} - \Delta vessel\ inventory}$$

The cumulative MUF, from PUREX initial operation in 1955 through December 1992, was in excess of 400 kg plutonium. Such a value is a cause for concern regarding the potential inventory of plutonium remaining in the PUREX facility during transition activities, however, facility management has concluded that there is minimal hazard attributable to this MUF. The bases for this conclusion involve both the errors inherent in the MUF calculation itself, and the post-campaign flushes and inspections conducted to ensure plutonium build-up was not occurring within the plant.

a. **MUF Calculation Errors.** Each term of the MUF equation included errors inherent in its method of calculation.

"$\mathbf{input_{Rx\ code\ calculation}}$" This factor was considered the major source of error in the MUF calculations. Through the early seventies, the plutonium input calculation was always based upon the predicted value obtained from reactor computer code calculations. This, despite the fact that several independent analysis methods indicated that the predictive code was consistently overstating the plutonium input, giving rise to inaccurate accountability calculations, and erroneous increases in MUF.

Based on a new PUREX head end sampling/analysis method developed in 1959 (the plutonium ratio method) and data gathered between 1959 and 1961, the predictive reactor codes were adjusted *downward* in July 1961. Data comparisons and further sampling/analysis improvements led to another *downward* revision in June 1965. However, from 1965 to the shutdown to standby in 1972, these corrective measures were overwhelmed by further errors in the reactor predictive codes that were (apparently) extremely sensitive to the gradual increases in fuel element exposure that were phased in across this period.

For PUREX operations from 1983 through the interrupted shutdown in 1988, the input factor was calculated from analysis of samples of the head end (D-5) dissolved reactor fuel feed tank. However, it was recognized that the method of obtaining the sample, using a double air lift system, still introduced some (uncorrected) biases into the calculation. Cumulative yearly MUFs for this period were generally an order of magnitude smaller than those calculated using the reactor predictive codes.

"$\text{output}_{\text{to PFP}}$" The output stream to the Plutonium Finishing Plant (PFP) was the best-characterized term in the equation. The purity of the product, as well as the multiple sampling and analyses conducted and relatively tight inventory controls tended to minimize the associated error-band.

"$\text{waste}_{\text{rough sample only}}$" The characterization of the waste stream was rudimentary at best. Typically, only one sample of an [assumed] homogenous waste transfer was collected, and analysis of the sample was limited by its innate chemical characteristics. Errors associated with this term were mitigated by the fact that the amount of the product that ended up in the waste stream was usually a very small percentage of the total throughput.

"Δvessel inventory" Vessel inventories were difficult to calculate during a campaign due to their dynamic nature. However, at the end of a campaign, the usual procedure involved system flushes designed to clear the process system vessels to output streams containing less than 0.01% plutonium, and resupply them with clean chemicals. Thus, the primary error associated with this term involved in-line holdup, vessel heels, and those sumps and catchbasins external to the normal process path. On at least one occasion, in trying to determine the cause of a MUF in excess of 40 kg in 1969, all of the vessels, sumps, and catchbasins were flushed and inspected: total plutonium yield was less than 10% of the MUF, with two-thirds of the recovered material flushed from the process line (e.g., L- and N-Cell packages) and the remainder identified through equipment washdowns and sump drainout.

Overall, when the material inventory books were closed following a campaign or a tracking year, they were never re-adjusted. The corrections made to the predictive reactor codes in 1961 and 1965, for example, were not employed to recalculate MUFs from previous years. The cumulative value of unaccounted-for plutonium was therefore assumed to be well below the published value for each year.

b. Post-Campaign Flushes and Inspections.

In 1988, PUREX was shut down in mid-campaign. A cleanout run was made in 1990, during which the following flushes were performed: all solution vessels; the plutonium nitrate gloveboxes, the plutonium oxide (dry side) gloveboxes. Flush solutions were routed back to the head end (D-5) tank, with final flush sample values indicating no (minimal) holdup in the system. However, not all of the systems were flushed. For example: the N-cell package was not cleaned out in this run, although it was subsequently cleaned (dry) as an individual project; and the sump air jet system and the vessel vent system were not flushed (or inspected).

Hold-up in the PUREX line was expected to be minimal, due to the acid basis of the process itself. However, in 1969, concern over potential accumulation of plutonium led to several visual inspections in the PUREX Facility. The "bathtub" (C-5) tank was geometrically most likely to contain residual plutonium, but an internal video inspection identified only negligible retention of potential plutonium. The facility air tunnel was inspected via still photographs, and the N-cell ductwork was assayed, but no sludges and/or layup materials were identified. The only appreciable quantity of plutonium isolated was approximately 500 grams found in some sludge from E-Cell.

Based on the flushes and cleanups performed, and the results of the inspections, PUREX management has concluded that the amount of unaccounted-for plutonium potentially remaining within the facility is minimal. However, criticality concerns and exposure control are still prime safety functions associated with each project that will be performed during facility transition and D&D efforts.

5. **Future DNFSB Staff Actions:** Two potential issues remain:

- Although the final PUREX cleanout run was completed in 1990, accountability records show that *negative* MUFs were recorded in 1991 (4.35 kg) and 1992 (3.36 kg). Thus, plutonium was being "generated" in PUREX, despite facility management claims that unaccounted-for material in the complex is minimal. The Staff will investigate the source of these kilogram quantities of plutonium.

- Staff monitoring of criticality and exposure controls during transition and D&D activities will be necessary to ensure appropriate levels of worker safety are maintained.

REFERENCES

1. WHC Letter, D. G. Harlow to W. A. Peiffer, dated October 5, 1993: *Review of Plutonium (Pu) Material-Unaccounted-For (MUF) at PUREX and Technical Reconciliations*. [93:6054]

2. R. P. Corlew, ARH-1445 - *Plutonium Unaccounted For (MUF) at PUREX*, dated November 5, 1969. [93:6055]

3. L. H. Taylor, ARH-1429 - *Historical Plutonium MUF Trend Analysis - PUREX - January, 1965 to July, 1969*, dated October 1, 1969. [93:6056]

4. L. H. Taylor & L. F. Wirfs, RL-CAO-165 - *Historical Review - Plutonium Ratio Receipt Measurement - July 1960 through December 1964*, dated May 25, 1965. [93:6057]