

DEFENSE NUCLEAR FACILITIES SAFETY BOARD

August 9, 1994

MEMORANDUM FOR: G. W. Cunningham
COPIES: Board Members
FROM: William Von Holle
SUBJECT: Trip Report for Visit to Lawrence Livermore National Laboratory

1. **Purpose:** This report contains the highlights of the visit of staff member William Von Holle on July 13-14, 1994, to attend a review at Lawrence Livermore National Laboratory (LLNL) of Defense Nuclear Agency (DNA) Propellant Sensitivity Program, discuss the Plutonium Immobilization Program and plutonium residue treatment research, and obtain an update on a recent plutonium storage can bulging incident. Staff members, Andrew De La Paz and Charles Martin, and outside expert Joseph Leary, visited LLNL separately on July 18-21, 1994, to perform a general safety analysis and review the storage of nuclear materials.

2. **Summary:**

- a. LLNL is developing promising techniques for plutonium residue treatment, but Department of Energy (DOE) has stopped all funding for such activity pending outcome of the EIS for plutonium disposition.
- b. The recent bulging can incident at LLNL is still undergoing investigation, but they believe they have identified the causes at this time.
- c. LLNL has taken the lead role in the Plutonium Immobilization Project (ultimate disposition). Vitrification is the leading candidate, thus far, with the final recommendation to be made following completion of the project in 1995.

3. **Background:**

LLNL has a relatively small plutonium inventory and a small research and development program for processing plutonium. The Livermore plutonium facility had not been formally reviewed by the DNFSB staff when other larger facilities were reviewed in preparation for the Plutonium Storage report, DNFSB/TECH-1.

The recent bulging plutonium storage can and the Board's interest in National laboratory leadership in plutonium technology, including the need for background information for the R&D requirement for Board Recommendation 94-1, sparked renewed interest in the LLNL storage practices and research program.

3. Discussion:

a. Processing Plutonium for Stable Interim Storage:

Several workers in plutonium chemistry briefed the staff member on their activities in treatment options for unstable plutonium residues. The staff member contacted Dr. Mark Bronson, Ms. Karen Dodson, and Dr. John Wilder.

Livermore has several promising candidates for processing unstable plutonium residues to stable metal or oxide. Among those are the following:

1. Advanced Salt Scrub Reactor: This could be made into a semi-continuous operation to reduce the plutonium in pyrosalts to metal with high efficiency. The advantage this may have over oxygen sparge is the coincident removal of americium, which LLNL claims does not happen in oxygen sparge.
2. Pressure Cycled Calcination: This method pulses air into the calcination oven alternately with a vacuum to expose fresh surfaces for calcination. The pulsing increases the efficiency of the operation and minimizes the need for worker intrusion. This minimizes worker exposure and area contamination. According to LLNL personnel, it is quite simple and elegant and adaptable to most existing furnaces in the DOE complex.
3. Molten Salt Destruction: This technique converts most waste material including rubber into clean gases by oxidation in molten K_2NaCO_3 plus O_2 . It would be done in two stages for plutonium contaminated wastes: Pyrolysis and calcination at 700C, followed by organic destruction at 950C. Plutonium oxide is recovered from the salt. This could be used for treatment of all the combustibles in the inventory. LLNL has a demonstration plant working on non-radionuclide contaminated waste materials like rubber.

A tour of the processing experimental area in the Plutonium Facility included observation of much of the apparatus for the above processes. However, progress has been slow and is about to stop for lack of funding. All funding for experimental work has been stopped until the EIS for plutonium disposition is completed.

b. The DOE Plutonium Disposition Project:

The LLNL program manager for SNM Disposition, Dr. Jeffrey Kass, and the program manager for Plutonium Immobilization, Dr. Leonard Gray, provided a description of LLNL's role in these activities. This project is to decide on the long-term form for storage (50 yrs.+) for plutonium as it comes from excess weapons and from process plants as oxide or metal. The Immobilization Project is roughly a two-year, \$9M, effort among the DOE facilities and some universities and institutes. The plan is to screen out most of the alternatives by September 1994, concentrating on the remaining two or three most promising concepts. The final choice will be made by September 1995. Vitrification is the leading candidate, according to the project leader. LLNL is scheduled to do a number of technical and economic assessments as paper studies. Only very limited funds are available for experimental or testing work.

c. Bulging Plutonium Oxide Cans:

During a tour of the LLNL Plutonium facility, the staff member observed two bulging Pu cans written up in a recent occurrence report, Report # SAN--LLNL-LLNL-1994-0040. A more recent radiographic examination of 15 cans found two with inner bulging cans. The cans contained impure oxide with 4% to 20% plutonium by weight; it consisted of a mixture of chem wipes, residues from the analytical laboratories and plutonium oxalate from solution recovery. It was reported to have been heat treated to 650 C, but not stabilized. Apparently this treatment was not sufficient to prevent subsequent radiolytic reactions, and the contents pressurized its sealed container. The bulging of the outer container would follow the leak or rupture of the inner can. All similar cans are scheduled to be opened and repackaged with material calcined at 800 C. It is interesting to note that this material was repackaged approximately one year ago.

Dr. Ralph Condit, investigator for the recent plutonium oxide bulging storage cans, updated the staff member on plans for the continuing investigation. LLNL personnel are planning to sample the pressurizing gases while opening the bulged cans as well as characterize the impure oxide material in order to establish the source of the gas.

d. LLNL Safety Surveillance Program:

Outside expert Joseph Leary joined DNFSB Staff members Andrew De La Paz and Charles Martin for a safety review visit the week following the first DNFSB Staff visit.

LLNL personnel perform nuclear material surveillance consistent with material control and accountability requirements. This includes bi-monthly inspections of all outer containers and selections of a small sample for nuclear material type and mass verification. However,

it was not evident to the DNFSB Staff that weight measurement data and packaging details (if an item was repackaged) were being provided to the appropriate LLNL personnel as part of a safety surveillance program.

e. The DNA Propellant Sensitivity Program Meeting:

The trip included a day-long review of DNA's Propellant Sensitivity Program, which has begun to systematically investigate various rocket propellant hazards for their ongoing weapon safety investigation . The DNFSB Staff member attended the review as an observer.

4. Future Activities:

The DNFSB Staff will continue to follow the progress of the bulging cans incident. Another visit is planned soon to discuss this and the Pu materials storage safety and accountability. The Staff will continue to monitor the interim stabilization and long-term disposition projects for application to Recommendation 94-1.