

DEFENSE NUCLEAR FACILITIES SAFETY BOARD

June 7, 1996

MEMORANDUM: G.W. Cunningham, Technical Director

COPIES: Board Members

FROM: Larry Zull

SUBJECT: Trip Report - Safety of Cesium and Strontium Capsules at Hanford

1. **Purpose:** This trip report documents a May 21-23, 1996, visit by the Defense Nuclear Facility Safety Board's (Board) staff to review the safety of cesium and strontium (Cs/Sr) capsules and materials at the Waste Encapsulation and Storage Facility (WESF) and Pacific Northwest National Laboratory (PNNL) Buildings 324 and 327 at the Hanford site. The review was performed by Board staff members Richard Tontodonato, Roger Zavadoski, and Larry Zull.
2. **Summary:** The review focused on WESF capabilities to safely store about 1900 cesium and strontium capsules in water-filled pool cells. The facility is in transition, planning for the decoupling from B-Plant support services October 1998, and upgrading some systems for continued safe storage of the capsules. However, a plan to decide the long-term disposition of the capsules, and hence the future of WESF, is not expected to be issued before December 1997.

An Interim Safety Basis (ISB), describing the current mission of safe storage and surveillance of the capsules, is to be submitted to the Department of Energy's Richland Operations Office (DOE-RL) for review by September 1996. A review of the original WESF seismic analysis, being conducted by ICF Kaiser Hanford Company, is scheduled to be completed by August 1996. A Safety Analysis Report (SAR) meeting the requirements of Department of Energy (DOE) Order 5480.23 is scheduled to be submitted to DOE for approval by October 1997. The Board staff intends to review these authorization basis documents when available.

Some capsules that were leased to commercial irradiators may have experienced chloride-induced stress corrosion cracking near the outer capsule welds due to a lack of water chemistry requirements and control. The current surveillance method does not detect degradation of the outer capsule prior to failure. Recognizing the need for more rigorous surveillance, the Westinghouse Hanford Company (WHC) requested a contractor to provide recommendations for improvements by September 1996. However, no date has been established for a WHC decision on the recommendations.

PNNL Buildings 324 and 327 contain about 700,000 curies of cesium material in the form of capsules, powder and pellets. Building 324 also contains about 8.3 million curies in the form of 35 vitrified canisters of cesium and strontium materials belonging to the Federal Republic of Germany. Buildings 324 and 327 are not seismically designed; however, the capsules and cesium materials are scheduled to be reencapsulated and sent to WESF before deactivation of the buildings is completed in October 1998. The vitrified waste will not be sent to WESF.

3. **Background:** From 1968 to 1985, WESF produced cesium chloride and strontium fluoride capsules which were stored in water filled storage pools at the facility. In the mid-1980's, 780 cesium capsules were leased to commercial facilities as gamma radiation sources. In 1988, after one leased capsule leaked radioactivity at a commercial facility, DOE began the recall of the capsules. All but 25 cesium capsules leased to a commercial irradiator and 13 cesium capsules and materials at PNNL facilities have been returned to the WESF storage pool. DOE has not determined the long-term disposition of the capsules (about 1300 cesium capsules and 600 strontium capsules), and the capsules will remain in the WESF pool cells for the indefinite future.

The Board's staff previously reviewed B-Plant/WESF during a May 15-18, 1995, site visit. The staff identified deficiencies with the safety basis, operations, and training, which the Board transmitted to DOE's Office of Environmental Management (DOE-EM) by letter dated August 3, 1995. Current WESF activities include safety upgrades, cleanout of the hot cells, and decoupling of WESF from B-Plant support systems. In October 1995, B-Plant began a three year deactivation program. The present review of WESF focused on the authorization basis, capsule integrity, capsule surveillance program, water chemistry, and exhaust ventilation systems.

4. **Discussion:** The Board's staff toured WESF and PNNL Buildings 324 and 327, and discussed safety related issues with DOE Richland Operations Office (DOE-RL), WHC, and PNNL personnel. The staff's observations are summarized below.
 - a. **Authorization Basis:** WHC stated that WESF, completed in 1974, was designed to withstand a Design Basis Earthquake (DBE) of 0.25 g horizontal acceleration in the pool cell area. However, ICF Kaiser Hanford Company has been reviewing the original seismic analysis as part of the development of an Interim Safety Basis (ISB) for WESF. The seismic review will be documented in a Natural Phenomena Hazard Survey report to be completed by August 1996. The ISB, which is to be completed by September 1996, will reflect the current mission of safe storage and surveillance of the Cs/Sr capsules. A Safety Analysis Report satisfying the requirements of DOE Order 5480.23, Safety Analysis Reports, is scheduled to be submitted to DOE for approval by October 1997. The staff intends to review the ICF Kaiser report, and the ISB, when available.

WESF is implementing a decontamination program which includes the cleanout of an exhaust duct in the K-3 Exhaust Ventilation System containing an estimated 20,000 curies of cesium and strontium materials from the hot cells. WHC plans to obtain samples to identify the isotopic composition and total curie content of the material. The role this material could play in a postulated accident is not clear.

- b. **Capsule Integrity and Surveillance:** The staff reviewed capsule fabrication, degradation, failure mechanisms, and the capsule surveillance program.
 1. **Capsule Fabrication** - The cesium chloride is double-encapsulated in 316L stainless steel tubes with welded end caps. One end cap was welded onto each tube before it was introduced to the WESF hot cells. These welds were radiographed to check their quality. The closure welds were made in the hot cells. No post weld heat treatments were performed. Both

inner and outer tubes were helium leak tested after closure welding, but only the outer closure weld was ultrasonically inspected. Due to problems in passing the ultrasonic inspection, the acceptance criteria were made progressively more lenient, eventually requiring that the weld only penetrate through at least 55% of the wall thickness. Furthermore, the DOE Type B investigation into the capsule failure at a commercial irradiator concluded that the ultrasonic inspection had substantial operator dependence as well as repeatability problems, with 95% confidence intervals of over $\pm 40\%$ of the predicted depth of penetration.

The strontium fluoride capsules were fabricated and inspected in the same manner as the cesium capsules, with the exception that the inner tubes were made of Hastelloy C-276, an alloy with better resistance to corrosion by fluoride compounds.

2. **Known Capsule Failures** - Capsule failures and other known defects fall into three categories: swelling, cracking, and weld flaws.
 - a. Swelling: Pure cesium chloride undergoes a phase change at 469o C that decreases its density by 15%. The material melts at 645o C with an additional 9% density decrease. Impurities can reduce the phase change temperatures significantly; 3% FeCl₃ allows melting to begin at 270o C. Impurity levels in the capsules are not documented, but testing of several capsules found sufficient impurities (e.g., >3% FeCl₃) to cause significant depression of phase change temperatures. Strontium fluoride does not have significant volume changes below 1050o C.

Although there is sufficient volume inside the inner capsules to accommodate the cesium chloride should it melt, swelling due to phase changes has been identified by DOE and WHC as the cause of the failure at the commercial irradiator. WHC believes that repeated overheating in air caused repeated phase changes that crumbled the cesium chloride, causing it to become packed in the bottom of the capsule. The inner capsule bulged, and eventually the bottom end cap weld ruptured. No defect was ever found in the outer capsule, even though it leaked. WHC believes the cesium escaped through interconnected microporosity or along stringers opened up by the strain imposed by the swollen inner capsule.

The remaining 11 cesium capsules which exhibited swelling were identified using a test method known as the clunk test (if sufficiently swollen, the inner capsule will no longer rattle inside the outer capsule when shaken). None of these capsules have leaked, and all were overpacked in welded stainless steel containers by PNNL.

- b. Cracking: One cesium capsule suffered a throughwall crack along about 45o of the circumference of the outer closure weld. This crack was discovered after operators reported a sluggish response in the

clunk test. The cause and age of the crack are unknown. WHC plans to perform a destructive examination of the capsule in FY96. The inner capsule has been extracted from the defective outer capsule and currently resides in a hot cell at WESF. No degradation of the inner capsule has been observed.

Weld Flaw: The last identified flaw is a single large pit found in one of the end cap welds of a cesium capsule. This flaw appears to be a fabrication defect and was found by a visual inspection after the capsule was received at WESF from a commercial irradiator. This capsule did not leak but is currently stored in a WESF hot cell as a precaution.

3. ***Other capsule degradation mechanisms*** - Other capsule degradation mechanisms have not been observed at WESF. However, since surveillance is limited to the clunk test, capsules which are degrading but have not yet failed would not be identified. The visual inspection performed on capsules returned by commercial vendors provides assurance that gross defects are not present, but would not identify fine cracks. Potentially significant degradation mechanisms are discussed below.
 - a. Stress corrosion cracking: The low chloride ion concentration in the WESF pools is expected to preclude stress corrosion cracking of the 316L outer capsules; however, commercial irradiators were not given any water chemistry guidance by DOE. State and Nuclear Regulatory Commission (NRC) operating licenses identified water conductivity requirements, but the chloride content of the commercial pools is unknown. Therefore, capsules leased to these irradiators may have experienced some degree of cracking, particularly in the areas of high residual stress and susceptible material condition near the end cap welds.
 - b. Fatigue: PNNL modeled fatigue crack growth in cylinder material and concluded that 35,000 thermal transients (removal from pool and equilibration in air) would result in only about 0.001 inch of crack extension. Since the worst-case capsules have experienced fewer than 10,000 such cycles, it appears that fatigue crack growth is not a significant degradation mechanism for the capsules.
 - c. Pitting: Pitting of 316L is unlikely in the low chloride, low conductivity water in the WESF pool. Conditions at commercial irradiators are less well-defined, but no pitting has been observed during receipt inspections of capsules returned to WESF.
 - d. Internal Corrosion: The inner capsule materials were specifically selected for their resistance to corrosion by the cesium and strontium salts. Limited empirical data, including inspection of the capsule that leaked at a commercial irradiator, indicates little internal corrosion is occurring.
4. ***Capsule Surveillance*** - Surveillance of the capsules at WESF is limited to quarterly clunk testing of the cesium capsules and monitoring the beta

activity in the pool. The strontium capsules were last clunk tested in 1989. WESF staff and management recognize that more rigorous surveillance is warranted. A subcontractor is evaluating enhancements and will provide recommendations to WHC by September 1996. No date has been established for a WHC decision on recommendations to DOE-RL on future surveillance methods.

WHC personnel stated that the principal enhancements being considered are a gamma scanner to monitor the cesium chloride configuration within the capsules, leak detectors that could monitor the water around isolated groups of capsules, and clunk testing the strontium capsules. The Board's staff believes that improvements are also needed in monitoring the condition of the outer capsules. Clunk testing and leak testing will not detect degradation of the outer capsule prior to failure. Considering that the capsules will be kept in the WESF pool for the indefinite future, and that two outer capsules have failed for reasons that are not clear, a more rigorous program for checking for external degradation appears to be needed.

In regard to water chemistry monitoring, commercial experience has found that some organic solvents, like trichlorethylene, will not give an indication (as increased conductivity or chlorides) of being present until they become dissociated by either thermal or radiolytic means. The commercial nuclear industry has increased its surveillance requirements to include total organic carbon measurements as well as to administratively control the types of organic materials that can be brought into pool areas. However, total organic carbon is not monitored in the WESF pool.

WHC is performing a survey of commercial experience to revise the WESF pool cell water chemistry surveillance requirements. When available, the Board staff intends to review the revised requirements.

- c. ***PNNL Buildings 324 and 327:*** Building 324, the Chemical Engineering Building, includes hot cells which contain a total of 7.7 kg of material in the form of cesium chloride powder and pellets. Also, about 34 canisters containing cesium and strontium material (8.3 million curies total) vitrified for the Federal Republic of Germany are currently stored in shipping casks in Building 324. Due to public protest in Germany, this material cannot be shipped.

Building 327, the Post-Irradiation Testing Laboratory, contains hot cells and a water storage pool. The eleven cesium capsules that failed the clunk test at commercial irradiators, and one non-swollen inner capsule, have been overpacked in welded stainless steel containers and stored in the Building 327 water storage pool. A thirteenth capsule with no known problems is stored in the same pool inside a mechanically sealed overpack container. To date, the overpacked capsules have all passed a monthly clunk test. Buildings 324 and 327 were not seismically designed. In addition, the buildings in the 300 Area are adjacent to private business parks and much closer to populated areas in

Richland than is WESF in the 200 Area. However, the buildings are scheduled to transition to EM-60 to begin a two-year deactivation program in October 1996.

The capsules in the Building 327 pool are planned to be cut open and the material repackaged in standard WESF capsules using equipment in the Building 327 hot cells. The other cesium materials are also to be processed and repackaged in standard WESF capsules. The new capsules are then to be transferred to the WESF pool for long-term storage. Before the deactivation of the building can be completed, the 34 vitrified canisters of cesium and strontium materials will be removed.

5. **Future Planned Activities:** The Board's staff plans to perform additional reviews of WESF, including structural/seismic integrity, authorization basis documents, and the capsule surveillance program.