Nuclear Forensics at PNNL:

A Case Study

Andrzej (Andy) T. Luksic

**Jon M. Schwantes, Matthew Douglas,
Steven E. Bonde, James D. Briggs, Orville T. Farmer
Lawrence R. Greenwood, Elwood A. Lepel,
Christopher R. Orton, John F. Wacker**

Pacific Northwest
NATIONAL LABORATORY

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

- Discovery of Material
- Field Analysis
- Laboratory Measurements
- Evaluation of Laboratory Data
- Additional Information
- Attribution

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

- December 2004: Washington Closure Hanford (WCH) was performing remediation of a waste site north of the 300 Area

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

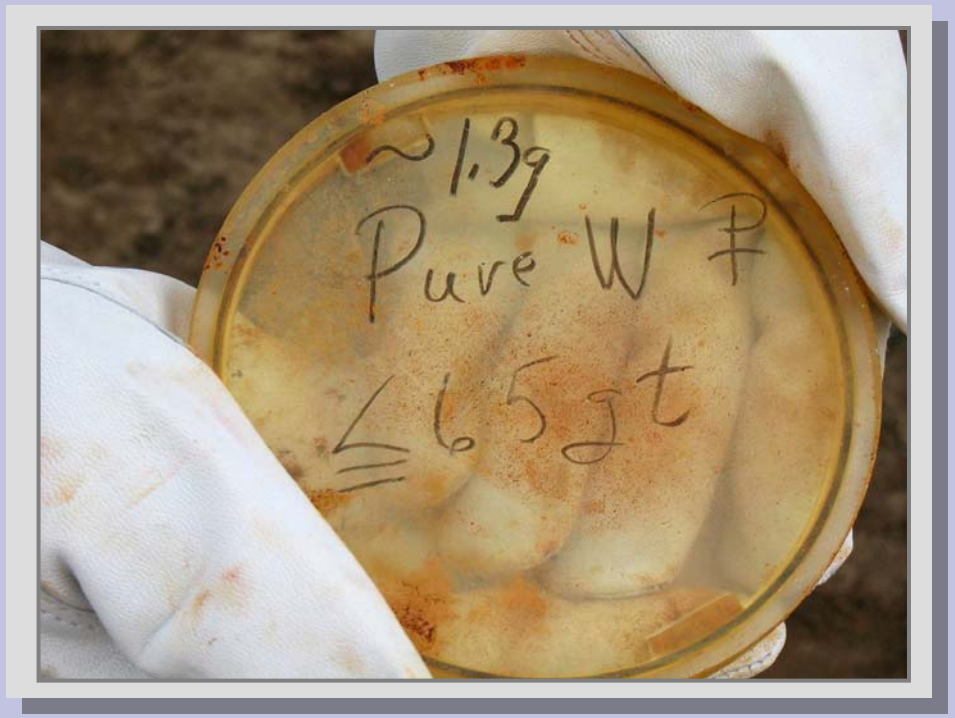
WCH uncovered a safe

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

The contents of the safe were examined

Nuclear Forensics at PNNL



Inside the safe was a surprise!

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

- *"LaF_3 for Recovery…"* written on bottle
- Preliminary field gamma analysis detected only ^{239}Pu (i.e., separated weapons-grade Pu)
- Limited paper trail indicated…
  - …safe sealed in 1945 after contamination
  - …disposed in early 1950's
- WCH estimated quantity of plutonium was < 1 gram

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

- WCH contacted PNNL for help in further characterization of the waste
- We were keenly interested because we had not identified any samples of Hanford plutonium, produced by irradiation of natural (virgin) uranium
- We wanted high-quality isotopic measurements
  - Pu-238 impacted by the use recycled uranium and enriched uranium
  - Pu-242 indicator of reactor type

Nuclear Forensics at PNNL

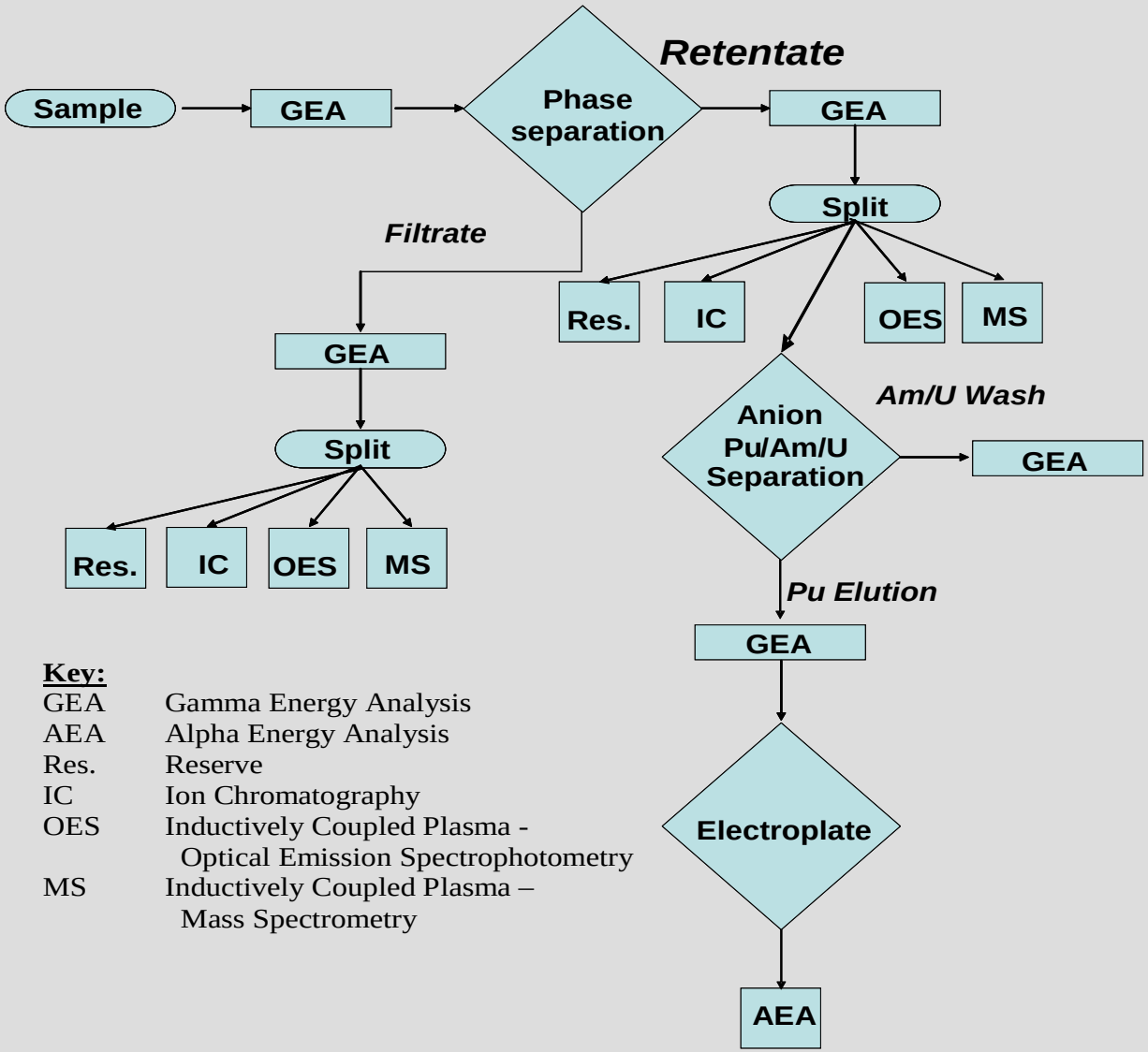
Pacific Northwest
NATIONAL LABORATORY

- The glass bottle contained ~400 milli-litres of solution and “crust” on the bottle walls
- PNNL obtained ~1/2 of the solution

“Jug Sample”

- Radio-chemistry was performed to determine isotopic composition of the contents and estimate quantity

Nuclear Forensics at PNNL



Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

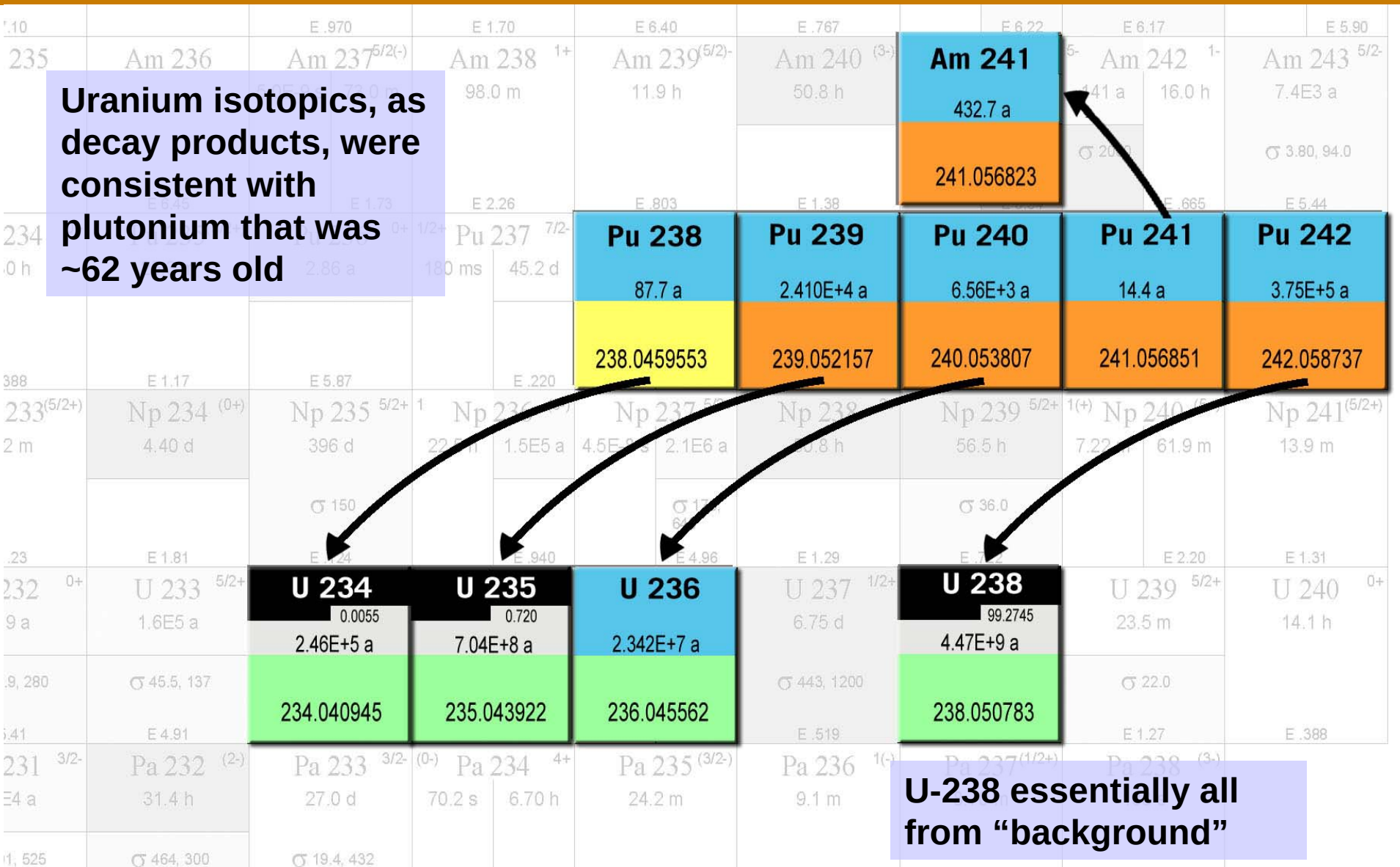
Pu-239	**99.961 w/o**
Pu-240	**0.039 w/o**

- This was extremely low-burnup plutonium, and probably represented some of the earliest Hanford-produced plutonium.
- We were very excited!

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

Uranium isotopics, as decay products, were consistent with plutonium that was ~62 years old



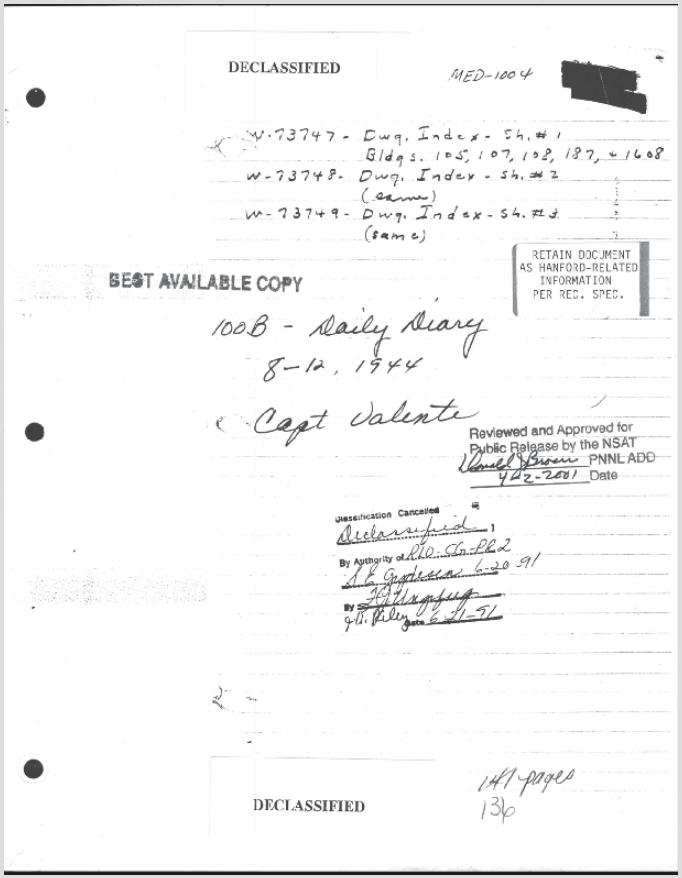
U-238 essentially all from “background”

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

- The first production dissolution at T-Plant began December 24th, 1944, of spent fuel discharge from B-Reactor beginning November 25th, 1944.
- Based on reactor operating records, the plutonium should have been slightly less than 1 w/o Pu-240
  - The “Jug Sample” had significantly less Pu-240
- So we started digging further …….

Nuclear Forensics at PNNL



DECLASSIFIED

MED-1004

W-73747 - Dwg. Index - Sh. # 1
Bldgs. 105, 107, 108, 187, & 1608
W-73748 - Dwg. Index - Sh. # 2
(same)
W-73749 - Dwg. Index - Sh. # 3
(same)

RETAIN DOCUMENT AS HANFORD-RELATED INFORMATION PER REC. SPEC.

BEST AVAILABLE COPY

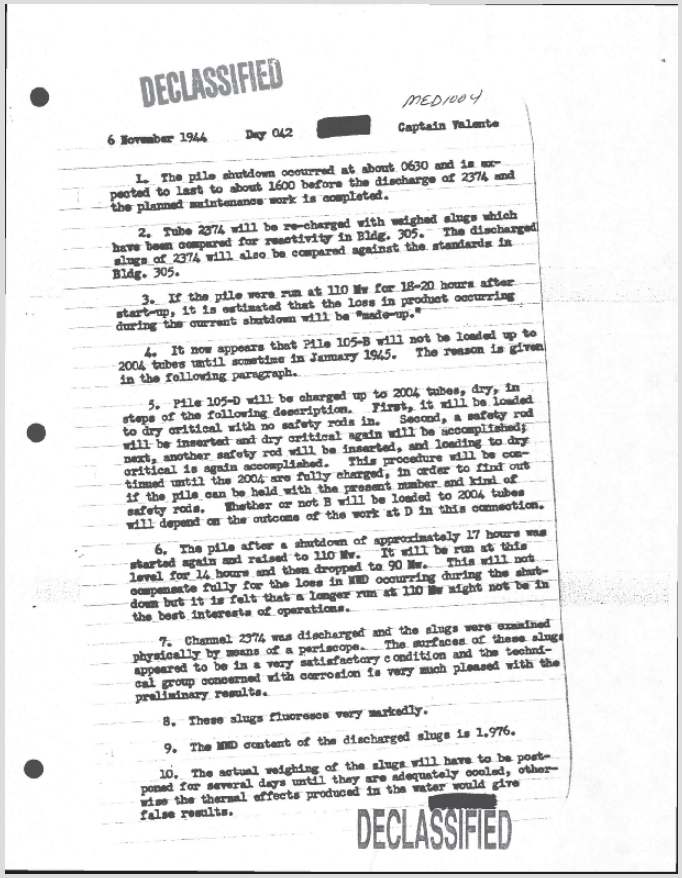
100B - Daily Diary
8-12, 1944
Capt Valente

Reviewed and Approved for Public Release by the NSAT
PNNL ADD
4-2-2001 Date

Classification Cancelled
Declassified
By Authority of RLO Sec Ltr 2
6-20-91
By
Date 6-21-91

141 pages
130

DECLASSIFIED



DECLASSIFIED

MED1004

6 November 1944 Day 042 [redacted] Captain Valente

1. The pile shutdown occurred at about 0630 and is expected to last to about 1600 before the discharge of 2374 and the planned maintenance work is completed.

2. Tube 2374 will be re-charged with weighed slugs which have been compared for reactivity in Bldg. 305. The discharged slugs of 2374 will also be compared against the standards in Bldg. 305.

3. If the pile were run at 110 Mw for 18-20 hours after start-up, it is estimated that the loss in product occurring during the current shutdown will be "made-up."

4. It now appears that Pile 105-B will not be loaded up to 2004 tubes until sometime in January 1945. The reason is given in the following paragraph.

5. Pile 105-D will be charged up to 2004 tubes, dry, in steps of the following description. First, it will be loaded to dry critical with no safety rods in. Second, a safety rod will be inserted and dry critical again will be accomplished; next, another safety rod will be inserted, and loading to dry critical is again accomplished. This procedure will be continued until the 2004 are fully charged, in order to find out if the pile can be held with the present number and kind of safety rods. Whether or not B will be loaded to 2004 tubes will depend on the outcome of the work at D in this connection.

6. The pile after a shutdown of approximately 17 hours was started again and raised to 110 Mw. It will be run at this level for 14 hours and then dropped to 90 Mw. This will not compensate fully for the loss in MWD occurring during the shutdown but it is felt that a longer run at 110 Mw might not be in the best interests of operations.

7. Channel 2374 was discharged and the slugs were examined physically by means of a periscope. The surfaces of these slugs appeared to be in a very satisfactory condition and the technical group concerned with corrosion is very much pleased with the preliminary results.

8. These slugs fluoresce very markedly.

9. The MWD content of the discharged slugs is 1.976.

10. The actual weighing of the slugs will have to be postponed for several days until they are adequately cooled, otherwise the thermal effects produced in the water would give false results.

[redacted]

DECLASSIFIED

Channel 2374 was discharged on November 6th, 1944

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

- Reactor calculations estimated the plutonium discharged from channel 2374 to contain ~0.13 w/o Pu-240
- “Jug Sample” burnup too low for B
- Safe contamination date preceded D, F, H discharges
- 305 test reactor power level too low
- Too much Pu-240 for production in accelerator
  - Seaborg, et al

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

- CP-1, CP-2, CP-3
  - Met Lab in Chicago
- Clementine, LOPO, HYPO
  - Los Alamos reactors
- X-10 (Clinton Works in Oak Ridge)
  - Initial criticality, Nov.4th, 1943
  - 1st recovered plutonium went to the Met Lab
  - 2nd recovered plutonium (?) went to Los Alamos

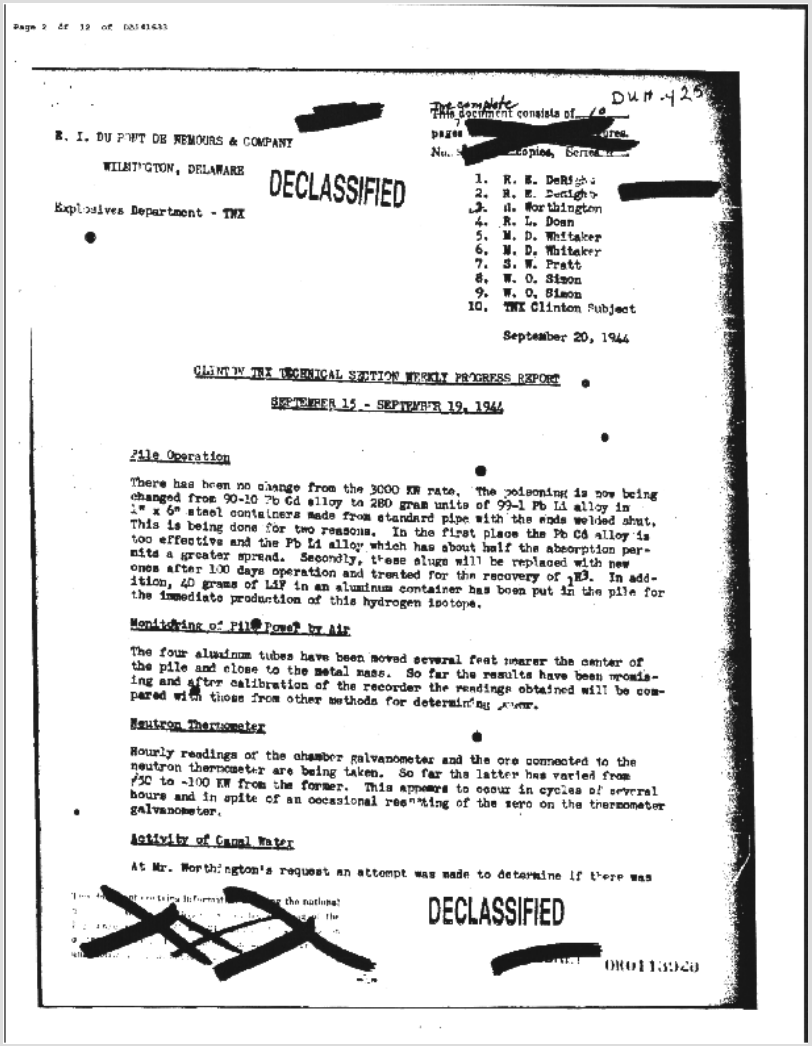
Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY

- As we worked to narrow down the possibilities, we found reference to a “hot” run in T-Plant prior to the “first” dissolution Christmas Eve, 1944
- As we dug deeper, we found:
  - mention of a large radioactive shipment from Clinton Works
  - A July 1944 request from Col. Matthias to Major Murphy, the District Engineer at Oak Ridge, for 100-day exposure material with 20 day cooling
- We eventually found the following notation in a Clinton Works weekly progress report ……

Nuclear Forensics at PNNL

Pacific Northwest NATIONAL LABORATORY



Page 2 of 12 of DA141633

DUM-425

E. I. DU PONT DE NEMOURS & COMPANY

WILMINGTON, DELAWARE

Explosives Department - TNX

DECLASSIFIED

This document consists of 12 pages. No. 3 of 10 copies, Series A.

1. R. E. DeRight
2. R. E. DeRight
3. H. Worthington
4. R. L. Doan
5. M. D. Whitaker
6. M. D. Whitaker
7. S. W. Pratt
8. W. O. Simon
9. W. O. Simon
10. TNX Clinton Subject

September 20, 1944

CLINTON TNX TECHNICAL SECTION WEEKLY PROGRESS REPORT

SEPTEMBER 15 - SEPTEMBER 19, 1944

Pile Operation

There has been no change from the 3000 KW rate. The poisoning is now being changed from 90-10 Pb Cd alloy to 280 gram units of 99-1 Pb Li alloy in 1" x 6" steel containers made from standard pipe with the ends welded shut. This is being done for two reasons. In the first place the Pb Cd alloy is too effective and the Pb Li alloy which has about half the absorption permits a greater spread. Secondly, these slugs will be replaced with new ones after 100 days operation and treated for the recovery of H^3. In addition, 40 grams of LiF in an aluminum container has been put in the pile for the immediate production of this hydrogen isotope.

Monitoring of Pile Power by Air

The four aluminum tubes have been moved several feet nearer the center of the pile and close to the metal mass. So far the results have been promising and after calibration of the recorder the readings obtained will be compared with those from other methods for determining power.

Neutron Thermometer

Hourly readings of the chamber galvanometer and the ore connected to the neutron thermometer are being taken. So far the latter has varied from +30 to -100 KW from the former. This appears to occur in cycles of several hours and in spite of an occasional resetting of the zero on the thermometer galvanometer.

Activity of Canal Water

At Mr. Worthington's request an attempt was made to determine if there was

DECLASSIFIED

Page 5 of 12 of DA141633

DUM-425

DECLASSIFIED

standard. It is not yet known if this film can be applied to uranium. Further tests with an improved film are planned.

Monitoring of 205 Stack

Continued monitoring for some time after dissolving had stopped did not bring the iodine activity to zero. It is now planned to introduce air at a point near the stack and continue operation to determine if the retention is in the main duct work or in the sampling piping and equipment.

Metal for Hanford

The following data are available on this material which left Clinton on September 17 in six containers packed in units of two in wooden cases with extra lead shielding.

Date Charged	Cont. No.	Row No.	Position Factor	Slug Position in Row	Avg. Power Rate	No. Slugs	Mgs. Prod.
5-11-44	1	2962	.810	8 thru 23	1720 KW	16	63.5
5-11-44	2	3064	.810	8 thru 23	1720	16	62.0
5-11-44	3	3064	.810	24 thru 39	1720	16	63.5
5-11-44	4	0664	.810	8 thru 23	1720	16	62.0
5-11-44	5	0664	.810	24 thru 39	1720	16	63.5
5-11-44	6	2962	.810	24 thru 39	1720	16	65.0

Readings on Containers (mr/hr at surface)

Top	Side
530	130
580	125
530	125
540	115
600	110
530	140

The slight difference in product content from the same channel is due to small changes in position factor when the metal channels were shortened several weeks ago.

DECLASSIFIED

-3-

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

- 96 spent fuel slugs from X-10
- ~112 kg uranium
- irradiated to 0.0036 MWD/kgU

  $\Rightarrow$ ~400 milligm Pu @ 0.03 $^{w}/_{o}$ Pu-240

Nuclear Forensics at PNNL

Pacific Northwest
NATIONAL LABORATORY

- Discovery of an unknown sample
- Field analysis indicated “pure” Pu-239
- Laboratory measurements provided isotopics & elemental data
- Evaluation of laboratory data provided estimates of material age, reprocessing, and irradiation history
- Additional information uncovered shipment to Hanford
- Attribution required integrating all of the above

Questions?

Pacific Northwest
NATIONAL LABORATORY