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UC-23

Plutonium-239/Beryllium Neutron Source Surveillance and Testing

R. D. Madding, Jr. and R. E. Vallée

February 15, 1973

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MOUND LABORATORY
Miamisburg, Ohio 45342

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ABSTRACT

A series of Mound-fabricated ^{239}Pu-Be neutron sources was subjected to destructive and nondestructive tests.

It is concluded that it is not possible, using techniques available to Mound, to assure integrity of the inner container. It is practical to determine that the outer container is sealed, although the quality of the weld joint cannot be assured. All the sample sources with a stainless steel outer container successfully passed the Department of Transportation special tests for special form. Two of the sources with a Vega steel outer container failed this series of tests, one in an extensive fashion.

Further use of selected sources is felt to be possible with a low degree of risk if the sources are qualified by the results of a series of NDT tests, reasonable restrictions are placed on their use, and periodic dimension, alpha wipe, and leak checks are made. Sources with Vega steel outer containers should not be used. A higher degree of assurance would obviously be obtained by providing the sources with an outer container using currently available quality-controlled fabrication procedures.

It must be noted that the recommendations and conclusions in this report can be applied directly only to Mound sources, the only fabricator represented in the destructive analysis and special test phases.

BACKGROUND

Neutron source fabrication began at Mound Laboratory in the late 1940's with the fabrication of ^{210}Po-Be neutron sources which were sold through the Isotope Pool at Oak Ridge. In 1956, Mound began fabricating ^{239}Pu-Be neutron sources.

The fabrication method, developed at Mound Laboratory and used until September, 1960, was as follows (Reference Figure 1): A weighed pellet of plutonium (c) was placed in the beryllium cup (a) which was in turn placed in the tantalum case (b). The tapered tantalum plug (d) was driven in flush with the top of the case and then sealed by tungsten-inert gas welding.

The assembly was then placed on an alumina support in a Vicor vacuum chamber and induction-heated to initiate the reaction. Although plutonium melts below 650°C, the reaction did not start until the temperature approached the melting point of beryllium, 1278°C. The heat of reaction then carried the temperature to about 2000°C.

When the source was cool, it was removed from the chamber and checked for wipeable contamination. Although the sources usually had a wipe count of less than 500 counts/min, the inner containers were occasionally contaminated by their surroundings in the glovebox during removal. In this case the inner containers were decontaminated using various techniques until the surface wipe count was less than 500 counts/min. The source was then placed into an outer steel jacket. The thick end plug, normally containing a 10/32 threaded hole for handling, was welded in place. After final neutron calibration, the source was ready for use.

An incident in August, 1960, whereby a bulging Pu-Be neutron source (S/N M-218) ruptured violently as the outer case was being removed, prompted an inspection of all sources. All users of Mound Laboratory sources fabricated prior to August 31, 1960, were requested to visually inspect their sources and measure their physical dimensions. The source users included the military, the AEC, other government agencies, universities and colleges, and private industries such as shipbuilders, scientific laboratories, and oil companies. In May, 1961, the users were requested to return all the sources to Mound Laboratory for inspection, testing, and recanning. Of the 743 sources, 668 sources were returned for recanning. The categories of users of the sources not returned are presented in Table 1.

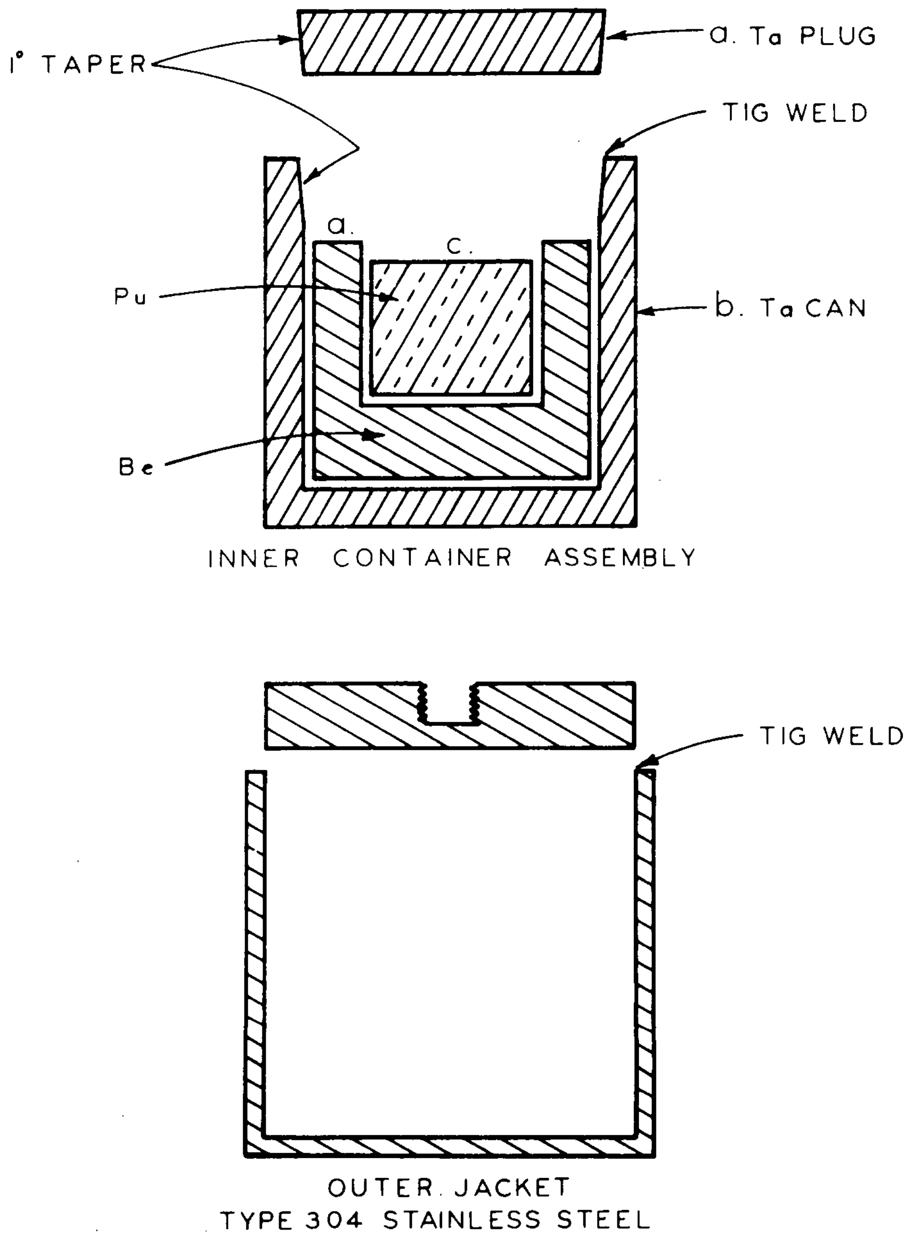

FIGURE 1 - Pu-Be source assembly.

Table 1

SOURCES SUBJECT TO RECANNING AND NOT RETURNED
BY USERS TO DECEMBER 31, 1963

Atomic Energy Commission Laboratories	18
Special U. S. Navy "Cable" Test Sources[a,b]	15
Other U. S. Government Agencies (Primarily Military)	8
Educational Institutions	7
Oil Well Logging Companies	13
Other Industrial Companies	2
Foreign Countries[b]	12
Total	75

[a] These are special, heavy wall, inspected to Naval reactor specifications.

[b] No attempt was made to have these returned.

The following operations were performed on the recalled sources:

a) A small hole was drilled through the center of one end of the outer container. This would furnish an indication of possible internal pressure, if it existed, and safely release it.

b) The outer container was machined off in a lathe.

c) The inner source container was helium leak checked. If a leak was found, the source was heated to drive out any possible trapped liquids.

d) If the inner container was leaking, it was rewelded and leak checked again.

e) The source was welded in a new outer container, leak checked, recalibrated, and returned to the user.

No sources other than source #M-218 (which prompted the investigation) had internal pressure. However, 142 sources had leaking inner containers. These 142 sources were recanned.

Immediately after experiments on the M-218 source, the fabrication procedure was modified to include a helium leak check of the inner container after reaction, and the sources were not immersed in liquids during decontamination. This modification precluded the possibility of internal pressure developing in future sources. It was also recommended that users measure the source diameters at maximum intervals of six months and report any increase in diameter >0.010 in. to Mound Laboratory.

Until last year, there was no attempt to evaluate the integrity of the sources in the field other than the inspections performed by the users as mentioned above. Mound Laboratory recommended to the AEC in March 1971 that a representative sample of the sources be recalled for dimensional, leak check, and radiographic inspection to evaluate the continuing integrity of the sources. It was also suggested that a sample of the sources undergo destructive analysis to determine actual fuel/metal compatibility and pressure increase with time. This recommendation was prompted by three instances of potential failure of neutron sources that were brought to Mound Laboratory's attention at that time. One source, returned to Mound by Schlumberger Well Services, Inc., had a wipe count on the surface of the source of 30,000 counts according to Schlumberger. This source was stored at Mound Laboratory in the logging tool in which it was used until its destructive analysis in May, 1972. A second source, received from NASA, Lewis Research Center, had an apparent bulge. Likewise, a source previously received from George Washington University had a noticeable bulge, and a radiograph of this source showed an apparent crack in the inner liner beneath the bulge. In addition, there were two returned sources which, because of use history, were not reused. There were in storage approximately 70 reuseable sources held for distribution to educational institutions under programs sponsored by the AEC Division of Nuclear Education and Training (DNET). The sources with the evidence of potential failure along with those held at Mound for distribution according to AEC/DNET were the ones recommended for nondestructive testing. It was also recommended that a small sample of sources undergo destructive tests. This original proposed program consisted of five phases as follows:

I. Examination of all sources in storage at Mound at the time.

II. Location of potential problem sources in the field by asking all licensees to perform tests on sources they hold and submit the data to Mound.

III. Recalling problem sources discovered in II above.

IV. Sampling the remaining sources in the field guided by any correlative factors derived in III.

V. Evaluation of all data and recommending future action.

The AEC authorized performance of Phases I and II, but only on those sources which were manufactured by Mound.

Efforts on the nondestructive testing and user (licensee) survey on Mound-fabricated sources began in April, 1971.

USER SURVEY

Letters with a survey form attached for each neutron source shipped (Exhibits 1 and 2) were sent to the last known holder of Mound-fabricated sources shipped by Mound. A total of 317 users of 1,226 ^{239}Pu-Be neutron sources were on record. The following basic information was requested:

a. wipe check
b. dimensional check
c. nature of use of source, past and present.

It should be noted that Mound is not and was not responsible for location inventory of these sources not at Mound (licensees are not required to notify Mound of shipment to another licensee). Additional information requested on the survey form included where the source was shipped to should the holder of record no longer have the source.

Approximately 50% of the users responded as follows:

Located		No Response Yet		Negative Response	
No. of Sources	Kg Pu-239	No. of Sources	Kg Pu-239	No. of Sources	Kg Pu-239
735	29.5	476	18.2	15	1.1

All negative responses except one represent return of the survey letters unopened due to firms having gone out of business or moved with no forwarding address now available, and some government activities no longer in existence. The exception was one firm whose records were archived and not readily accessible, with no current record of possessing the sources in question.

Interim guidance provided by the AEC in March 1972 directed that efforts regarding the survey be discontinued. (R. Roush, A. F. Schmidt, S. L. Snider)

NONDESTRUCTIVE TESTING

The nondestructive testing (NDT) program initially began on Mound sources, but was soon applied to all sources being made available under DNET when anomalies, particularly from radiographic inspection, became apparent in Mound-fabricated sources. Interim guidance was provided by the AEC in January 1972, which discontinued shipping of non-Mound-fabricated sources under DNET auspices due to possible viewing of this practice as competition with private industry. Additional guidance received on April 27, 1972, halted all shipments of neutron sources from Mound to users indefinitely.

August 9, 1971

ABC Drilling Inc.
300 508th Street
Denver, Colorado

Attention Mr. R. K. Roe

Dear Mr. Roe:

We at Mound Laboratory are in the process of taking a survey of the condition, disposition, and use of plutonium-239 beryllium neutron sources which were fabricated by us.

According to our records, you have in your possession the neutron sources listed on the enclosed printout. We would appreciate your verification of this list, and completion of an enclosed "Plutonium-239 Beryllium Neutron Source Survey" form for each source in your possession. Please return the completed form to us by September 15, 1971. In the event that you have transferred a source to another licensee or otherwise disposed of a source, please complete the appropriate sections of the form showing what the disposition of the source is and return the form to us.

If you have any questions concerning the information we request or the methods for taking the measurements, or if our records are incorrect or inconsistent with your records, please contact Mr. A. F. Schmidt at Mound Laboratory, telephone (513) 866-7444, extension 3172.

Very truly yours,

W. T. Cave

W. T. Cave, Director
Nuclear Operations

WTC:km
Enclosures

EXHIBIT 1

Source #M-__________

MONSANTO RESEARCH CORPORATION
Mound Laboratory
Miamisburg, Ohio 45342

PLUTONIUM-239 BERYLLIUM NEUTRON SOURCE SURVEY

GAUGING DATA:

Measure diameter (nearest 0.001") of source near top (serial # end), at center, and near bottom (opposite serial #).

Diameter	Measurement #1	Measurement #2 (at 90° to #1)
Top	__________	__________
Center	__________	__________
Bottom	__________	__________

WIPE TEST:

Wipe the source thoroughly with a 1" diameter piece of filter paper and check the paper for loose alpha contamination.

Result: __________ counts/minute alpha

VISUAL EXAMINATION:

Any noticeable defects or damage? ☐ Yes ☐ No

If yes, please state nature of defect or damage.

____

____

____

USE:

Please state nature of use to which the source is being or has been put.

____

____

____

LOCATION OF SOURCE:

Company: ____________________; City: __________; State: ________

OTHER INFORMATION (Check Appropriate Box):

- ☐ Currently leased from AEC
- ☐ Purchased (not leased)
- ☐ Procured through AEC Division of Nuclear Education and Training sponsored program agreement # NET__________
- ☐ Source not in our possession:
  Shipped to ______________________________
  ____
  on (date) ______________________________
- ☐ Source can be made available for temporary return to Mound Laboratory for inspection

____

Signature (Title) Phone Date

Please detach yellow copy for your file. Send original to:

Monsanto Research Corporation
Mound Laboratory
Miamisburg, Ohio 45342
Attn: A. F. Schmidt

EXHIBIT 2

The technique for each of the nondestructive tests is as follows:

Radiography Two radiographic views were made at 0° and 90° intervals, using an iridium-192 source and a two-speed film combination (type "M" film for the liner and type "SR" film for the outer container) in each exposure. Exposures ranged from 45 to 80 Ci-hr, depending on the size of the source being radiographed.

A high-intensity Keleket illuminator and 7X comparator were used for film reading, with reporting in the following areas: general condition of the outer container, integrity of the outer container weld, minimum weld penetration, fuel-liner corrosion, integrity of the inner weld and minimum weld penetration, and any abnormal condition detected in reading the radiographs. (M. I. Gray, J. Stockton)

Leak Check If both the inner liner and outer container exhibited no anomalies in radiography, a leak check was performed by first "soaking" the sources in a helium pressure vessel maintained at 300 psi for 30 min, then placing them in a vacuum chamber directly connected through the manifold to the spectrometer detector. This leak detector was calibrated using a calibrated standard helium leak and set up so that a direct readout of the actual leak rate could be made. This system would detect leaks of less than 1×10^{-6} std cc/sec of helium. (M. I. Gray, J. Stockton)

Visual-Dimensional Check If no anomalies were detected by radiography, the visual-dimensional check was performed. Equipment was borrowed from another program to establish the feasibility of its use on neutron sources, and all were not checked.

The Remote Measuring System (RMS), developed at Mound for another programmatic application, was used for the dimensional measurements. It consists of a large base approximately 4 ft long with a 1 ft square moveable plate mounted on top that moves the length of the base on two precision ball slides. This linear movement is monitored to the nearest 0.0001 in. by a Vernac readout system. A Brunson model 75 Transit Square is attached to the moveable plate. A Brunson alignment scope, model 83, with a built-in autocollimation unit is used to maintain the attitude of the axis of the Transit Square during the actual reading of the diameter of the source. A remote control rotary table is used in conjunction with the RMS to position the part and reduce the exposure. The Transit Square is approximately 15 ft from the source, and the alignment scope is approximately 15 ft from the Transit Square. Dimensional accuracy to ±0.001 in. is attainable. Length and diameter measurements were taken at various locations on each source.

Visual inspection was performed with the Transit Square at 30X and with a Questar telescope at 80X. The visual was used to confirm a bulged condition or any bad tool marks, scratches, cracks, or other flaws that were detected by the eye alone. (J. R. Marshall)

Dose Rate and Alpha Wipe Check A 10-in. Spherical Neutron Dosimeter (Texas Nuclear, series 9140) was used to determine the neutron count rate, and a Gamma Survey Meter (Victoreen 440) was used for the gamma emission rate from the bare neutron sources. The neutron/gamma rate was determined and recorded at varying distances in air. There was no attempt to control or compensate for the scattered radiation, except all objects were at least 5 ft from the source or instruments (walls, floor, or other surfaces). Measurements on each source were made under essentially identical conditions. All recorded data were plotted on 3 x 3 cycle log graph paper for inverse square law observations.

An alpha wipe check was performed on each source and counted in a scintillation alpha counter for any detectable alpha radiation. (L. G. Musen)

Neutron Emission Neutron emission measurements were obtained with a precision long counter by comparing the count rate from the unknown neutron source to the count rate from a standard source. The emission rate of the standard source had been calibrated by the National Bureau of Standards. Typical emission rates for the neutron sources have an uncertainty of ±3%. The relative uncertainty between any two measurements is normally ±1%. (R. A. Neff)

Results are tabulated in Table 2 for Mound-fabricated sources. Appendix A tabulates results from non-Mound-fabricated sources from the DNET loan program inventory. Column totals represent the maximum number of sources to which the column heading is applicable. Some sources exhibited more than one anomaly. Radiography was performed before leak check, and if sources were found to have open inner containers, leak checks were not performed due to the risk of contaminating expensive helium leak equipment and to these sources being deemed unsuitable for further use.

SPECIAL TESTS (DoT Tariff 25: 173.398a)

The NDT results raised a significant question regarding the suitability of having these sources in circulation. It was recognized that the destructive tests yet to be performed would not provide a satisfactory

Table 2

NONDESTRUCTIVE TEST SUMMARY

MOUND-FABRICATED SOURCES

Test	No Evidence for Potential Problem	Evidence for Potential Problem	Remarks	Tested
Radiography	20			53
		2	Outer liner open	
		3	Outer liner bulging	
		20	Inner liner possibly open	
		5	Inner liner apparently bulging	
		9	Apparent fuel-liner corrosion	
Leak Check	23			24
		1	Leak >3 x 10^{-6} cc/sec	
Visual-Dimensional	13			13
Dose Rate and Alpha Wipe	53			53
Neutron Emission	25			25
Totals	20[a]	33		53

[a] Controlled by radiography.

answer to this question. Design criteria and historical data against which NDT results could be meaningfully compared were not available. An engineering evaluation could not be made because of insufficient information being available regarding the materials used and fabrication of each source.

It was recommended to the AEC on March 3, 1972, that the Department of Transportation (DOT) special tests for special form materials (Tariff 25, paragraph 173.398a) be used as the test criteria to establish the suitability of the source design.

Parallel with this effort, the possible application of ultrasonic techniques to determination of weld penetration on the outer container was investigated. This technique was determined to be unsuitable due to the variation in the configuration of the joint areas of the hardware parts used in fabrication. (W. A. Dudley)

The integrity of the inner container cannot be assured with any degree of certainty using known techniques. Radiography does reveal the condition of the liner through one plane, but cannot be used to detect a defect which would allow a 1×10^{-6} std cc/sec leak of helium. Thus, an integral inner container could not be part of the design criteria.

Prior to initiation of the special tests, 12 Mound-fabricated sources were available which had been radiographed as part of the NDT program and in which the only defect exhibited was a possibly open or open liner. These had not been leak checked as part of NDT, but were leak checked for use in the special tests. Four of these sources, including M-436 which was then introduced into the destructive test phase, indicated leaks substantially greater than 1×10^{-6} std cc/sec. No alpha contamination was released from these sources. The leak was too great to permit use of the leak detector, and the helium bubble technique was employed first as a screening test, and later as the leak check between special form tests. This technique consists of the helium pressurization described in the NDT section, and immersion individually in a glass beaker of alcohol for at least 2 min. Leaks greater than 1×10^{-6} std cc/sec can be detected.

The additional 10 Mound-fabricated sources used in the special tests were obtained from a group of 12 sources recently returned to Mound from off-site users. Again, no alpha contamination was detected on the two sources exhibiting leaks. A leak rate determination (see the NDT section) and alpha wipe test only were performed on these 10 sources prior to the special tests (free drop, percussion, heating, and immersion) being performed on each source in sequence.

Free Drop Test In this test each source is free dropped from a distance of 30 ft onto a flat, essentially unyielding, horizontal surface, striking the surface in such a position expected to suffer maximum damage. Due to the potential release hazard (which was considered to be very low), the free drop was conducted in a radioactive control area (Alpha Fuels Building, Cell 112). Previous impact studies indicated that the angle of impact at which maximum damage would occur is 45° from vertical end on.

To minimize spread of radioactive contamination in the event of source rupture, a 1/2 in. steel plate was mounted inside a 77-gal drum supported by lead bricks. Extending into the drum were an air alpha monitor sample tube and a filtered vacuum cleaner hose to detect and remove, respectively, released radioactive material.

The apparatus, shown schematically in Figure 2, was designed, built, and assembled in Alpha Fuels Building, Cell 112. After the drop apparatus was checked, several test runs were made using dummy sources to verify the impact angle and drop area of the sources.

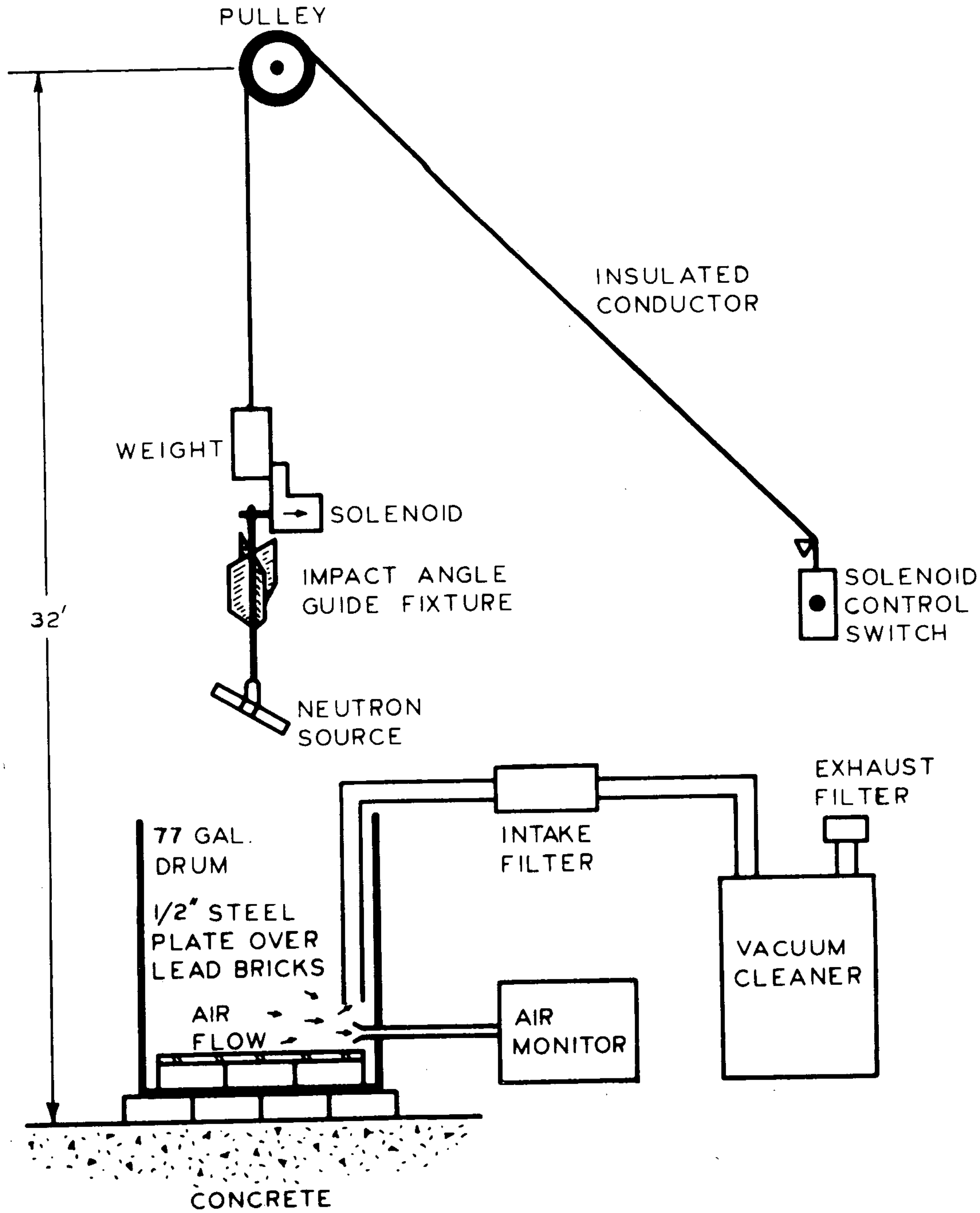

FIGURE 2 - Schematic of the free drop test apparatus.

The sources were removed one at a time from the transfer container, wiped for alpha contamination, and monitored for gamma and neutron emission.

The sources were then mounted one at a time on the solenoid-activated release mechanism and hoisted to a predetermined 30-ft level by pulling the electrical cord through a pulley. Once the source stabilized at the 30-ft level, the solenoid release mechanism was activated allowing the source to free drop 30 ft and impact the 1/2-in. steel plate at approximately a 45° angle inside the 77-gal drum. A wet towel was immediately draped over the source to prevent the spread of activity if the source had ruptured on impact. An alpha wipe was taken immediately after impact. No contamination was found. Each source was then helium pressure/bubble tested prior to percussion testing. (J. E. Selle, C. O. Brewer)

Percussion Test This test was performed in a Lucite box as shown in Figure 3. Each source was alpha wiped and placed on its side on a 1/4 in. thick sheet of lead which in turn was resting on a smooth 1/4 in. thick sheet of steel. A 3-lb, 1 in. diameter, steel rod was suspended 40 in. above the sample by a string extending through a hole in the top of the box and attached to a ring stand. The box was closed, the exhaust blower was started, and the weight suspension string was severed; the weight dropped on the source. The air monitor connected to the box was checked. If no evidence of alpha contamination was found, the lower box door was opened and an alpha wipe taken. Upon a negative result, a thorough alpha wipe check of the lower box interior was made. If again a negative alpha wipe resulted, the neutron source was stored for subsequent leak checking using the helium bubble technique.

Very minor, if any, physical change in the shape of the sources was observed with no evidence of alpha contamination or helium leak. (C. P. Johnston, D. R. Schaeffer)

Heating Test The sources, one or two at a time, were placed in an air-atmosphere muffle furnace (not sealed) located in a fume hood, and heated for a predetermined time to bring them to 1475°F. They were held at this temperature for 10 min. The sources were then removed from the furnace, placed on a fire brick in the hood, and alpha wiped with a glass fiber wipe. A negative result was followed by a wipe after they had cooled for approximately 5 min. Again a negative alpha wipe result was followed by cooling in air for about 30 min. The sources were then water cooled and an additional alpha wipe check made. It was negative in each case, and this was followed by helium bubble leak

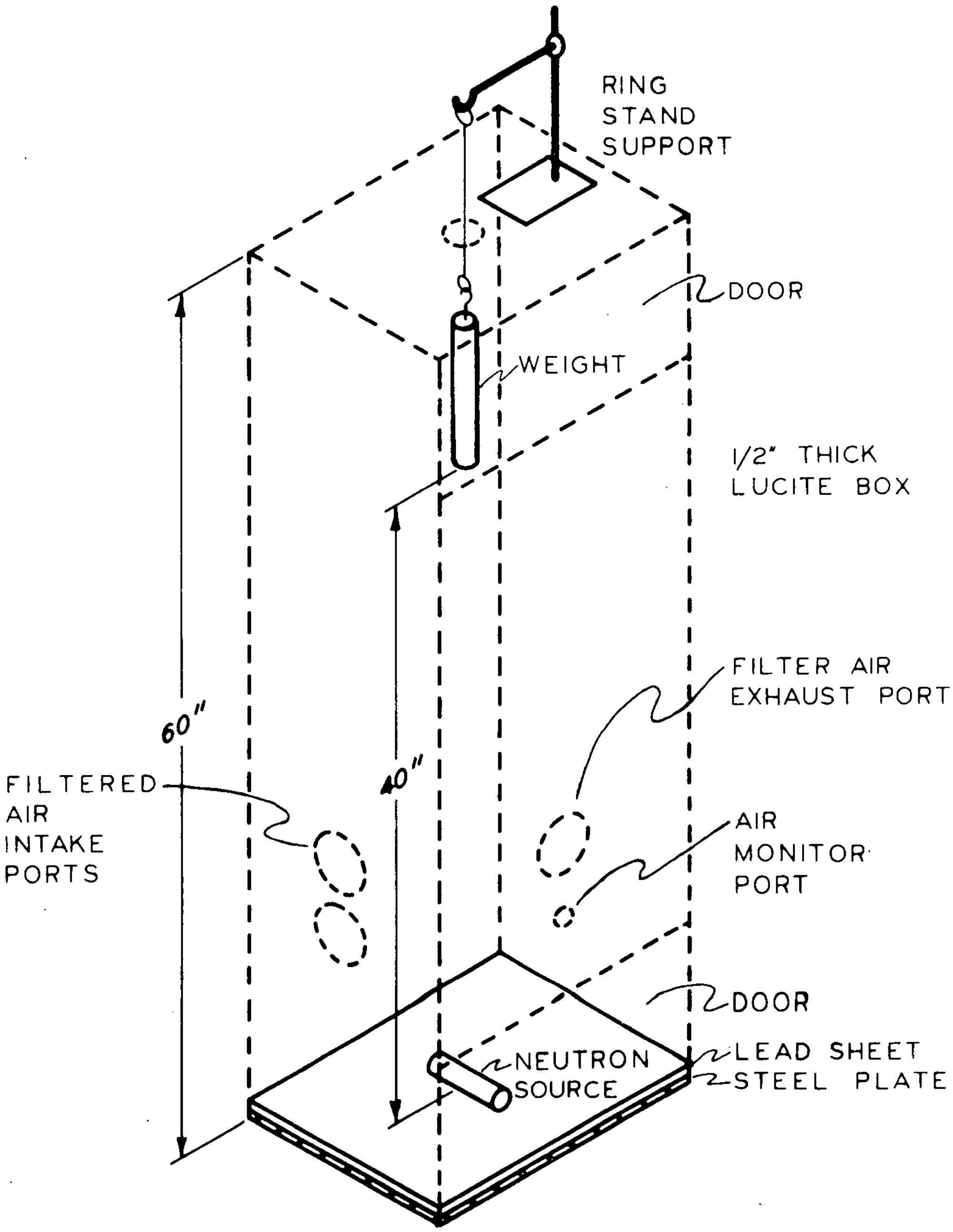

FIGURE 3 - Schematic of the percussion test apparatus.

checking and storage. No leaks were detected. The larger dimension sources exhibited substantial scaling (oxidation) as a result of the heating test. (C. P. Johnston, D. R. Schaeffer)

Immersion Test This test consisted of immersion for 24 hr in water at room temperature in clean plastic containers with lids. The water was at an initial pH of 6.5 with a conductivity of leas than 10 $\mu\Omega$/cm. At the end of the 24 hr the immersion water was sampled and analyzed for alpha contamination. The sources were removed from the containers, dried, and alpha wiped. Source M-1006 was found to have a crack on the bottom extending nearly the full length of one side with the inner container visible. Drying the source, wiping, and counting gave a result of approximately 1500 dis/min. Source M-1127, which was in the same immersion vessel with M-1006, was found to have a wipe count of about 500 dis/min. M-1127 was easily cleaned and placed in the storage container. Subsequent helium leak rate checking indicated that this source and 15 of the 16 other sources were not leaking. A leak in source M-1013 was found using the helium leak rate apparatus; it was not detectable by the helium bubble method.

Scintillation counting of the M-1006 immersion water samples gave no evidence of alpha contamination, indicating little or no solubility of the leaking material under the conditions of the experiment.

Sources M-1006 and 1013 were those returned from Schlumberger Well Services, Inc., and the outer container material had been specified by them as it had for M-1019, the source which they found to be leaking and returned to Mound in 1970. A complete discussion regarding the "Schlumberger" sources is incorporated in the following Destructive Test section. (C. P. Johnston, D. R. Schaeffer)

A tabulation of source data, nondestructive tests performed, and special test results is provided in Table 3.

Table 3

SOURCES SUBJECTED TO SPECIAL TESTS:
FREE DROP, PERCUSSION, HEATING, AND IMMERSION

Source No.	^{239}Pu (g)	Mfg. Date	Recan Date	Radio-graph	Pre-leak Test Rate	Post-leak Test Rate
M-253	79.74	9/22/58	9/27/62	-	O.K.	O.K.
M-258	79.90	12/17/58	7/17/61	x	O.K.	O.K.
M-273	79.52	12/17/58	7/17/61	x	O.K.	O.K.
M-275	79.88	12/17/58	8/24/61	x	O.K.	O.K.
M-471	79.63	4/20/59	7/29/63	x	O.K.	O.K.
M-475	8.00	5/28/59	9/14/62	-	O.K.	O.K.
M-618	79.98	2/8/60	7/17/63	-	O.K.	O.K.
M-762	79.65	6/16/60	5/19/61	-	O.K.	O.K.
M-873	160.05	1/27/61	-------	x	O.K.	O.K.
M-909	15.97	11/11/60	-------	-	O.K.	O.K.
M-912	11.99	12/20/60	-------	-	O.K.	O.K.
M-914	14.95	1/18/61	-------	x	O.K.	O.K.
M-923	15.98	11/18/60	-------	-	O.K.	O.K.
M-932	31.94	12/20/60	-------	x	O.K.	O.K.
M-950	63.99	1/18/61	-------	-	O.K.	O.K.
M-1006	78.83	7/27/61	-------	-	O.K.	[a]
M-1013	76.13	7/26/61	-------	-	O.K.	[b]
M-1127	75.40	-------	10/11/61	x	O.K.	O.K.

[a] Failed in immersion; not leak checked ("Schlumberger" source).
[b] Failed; leak rate >3 x 10^{-6} std cc/sec ("Schlumberger" source).

DESTRUCTIVE TESTS

The destructive test series consisted of the following:

a. Internal pressure measurement (gas tap), sampling, and analysis of the contained gases.

b. Metallographic analysis of the outer container, inner container, and fuel.

Ten sources were selected for destructive analysis. Nondestructive tests (NDT) were performed on these sources (reference Appendix A) prior to destructive analysis. Sources were selected which represented two areas of interest: first, those which had been subjected to unusual environments or which had visual defects, alpha wipe anomalies, or leaks. These include M-9, 71, 75, 436, and 1019. Sources M-493 and 1053-S were also of interest because of radiographic evidence of inner liner corrosion and possible fuel migration. Secondly, sources were selected for which no significant anomalies were noted in NDT. These included M-472, 1166, and 1190.

Since sources M-436 and 1019 were known to be leaking, they were not gas tapped. Sources M-493 and 619 were gas tapped only to obtain additional gas analysis data.

A table showing the nondestructive tests performed and a brief description of the history and predestruct condition of each source is given in Appendix B.

Internal Pressure Measurement, Sampling, and Analysis of the Contained Gases

Introduction Ten Pu-Be neutron sources were tapped for gas samples, some of which were lost or contaminated. Two of these, M-71 and 1053-S, were drilled through the flat end with extreme difficulty over a long span of time. The gas collection and pressure measurement technique was modified and the data from these two sources rejected. Data on M-9 exhibited internal pressure - volume contradictions which resulted in its rejection. The following describes the method of gas sampling and contains a compilation of the results of the successful tests.

Equipment The pressure sensing and gas sampling apparatus used in the studies consisted of a vacuum manifold to lower the system pressure, an MKS-type 77H-300 pressure sensing head connected to a MKS 100A series digital pressure readout to measure released gas pressure, a standard reference volume, a drill press, a vacuum chamber to hold the sources, and a Toepler pump to transfer the released gases to a sampling tube (see Figure 4). The entire apparatus was operated in a nitrogen atmosphere glovebox.

The drill bits used for opening the capsule were locked into a hardened, stainless steel extension and inserted into the opening chamber through a triple o-ring seal. The upper end of the extension was connected to a drill press.

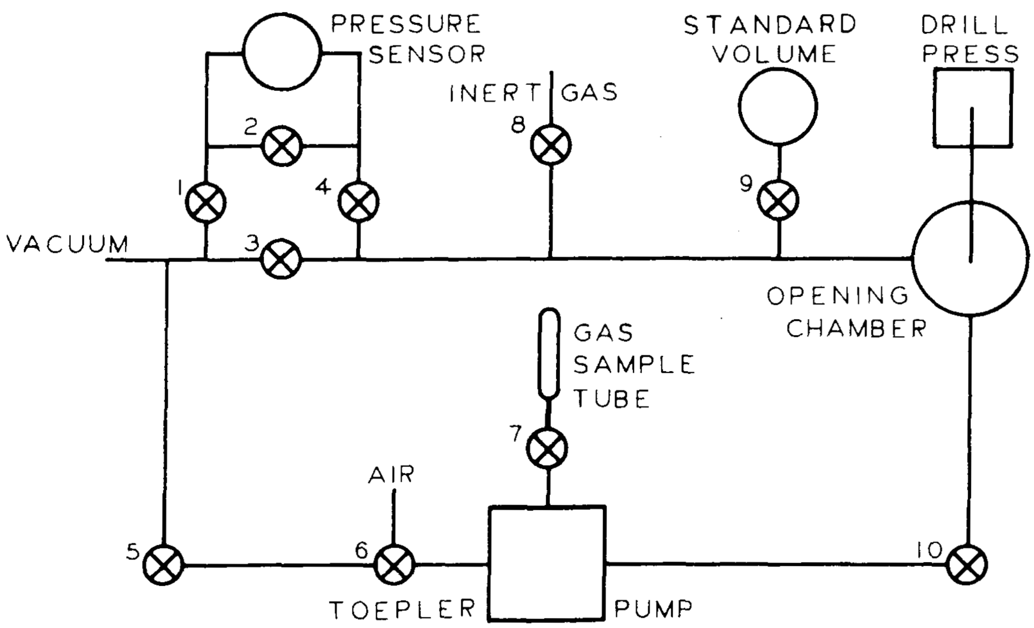

FIGURE 4 - Schematic of the gas tapping equipment.

Procedure An initial volume of the empty system was determined as follows:

a. The entire system was evacuated.

b. The system was backfilled with argon and its pressure noted.

c. The same pressure was trapped inside the standard volume and the rest of the system evacuated.

d. The gas in the standard volume was released into the system and the new pressure noted.

e. The system volume was calculated using $P_1 V_1 = P_2 V_2$ assuming a constant temperature.

This same method was used to find the system volume with the source in place and to find the free volume inside the source.

Volume of Source = (volume of system) - (volume of system and source)

$$V_S = V_{sys_1} - (V_2)$$

Free Volume of Source = (volume of system and source) - (volume of system and tapped source)

$$V_{SF} = (V_3) - (V_2)$$

Pressure on Standard Volume = P_1

Volume of Standard Volume = V_1 = 317.5 cc

System Pressure after Release From Standard Volume = P_2

Example:

<u>Volume of Source M-259</u>

P_1 = 243.1 mm
V_1 = 317.5 cc
P_2 = 69.5 mm
V_{sys_1} = 1180.3 cc
$P_1 V_1 = P_2 V_{sys} = 243.5 \times 317.5 = 69.5\ V_2$
V_2 = 1110.6 cc
$V_S = V_{sys_1} - V_2$ = 1180.3 cc - 1110.6 cc = 69.7 cc

<u>Free Volume in Source M-259 Inner Container</u>

P_1 = 243.5 mm
V_1 = 317.5 cc
P_2 = 68.9 mm
V_2 = 1110.6 cc
$P_1 V_1 = P_2 V_3 = 243.5 \times 317.5 = 68.9\ V_3$
V_3 = 1122.1 cc
V_{SF} = 1122.1 - 1110.6 = 11.5 cc

<u>Gas Pressure in M-259 Inner Container</u>

System Volume during Gas Tapping = (Volume of original system standard volume) - (Source Volume - Free Volume of Source)

$$V_{sys_2} = (V_{sys_1} - V_1) - (V_S - V_{SF})$$

Pressure of $System_2$ after tapping source M-259 = 1.14 mm

Free volume in source M-259 inner container = 11.5 cc

Gas pressure in inner container = P_I

$P_{sys_2} \times V_{sys_2} = P_I \times V_{SF} = 1.14$ mm $\times$ (1180.3-317.5) - (69.7-11.5) cc = $P_I \times 11.5$ cc

$P_I = 79.8$ mm = 1.5 psia

Discussion One source (M-1066) indicated such a slight gas pressure release that no gas analysis was taken.

The higher pressures inside of some sources may have been caused by the source fabrication technique of driving in the tapered end plugs and pressing the inner container into the outer container. This could compress any trapped gases.

Several of the sources gave only one pressure indication when they were tapped. There are two possible reasons for this: 1) the inner container wall was ruptured and all gases escaped when one container was tapped, 2) the proximity of the inner container wall to the outer container wall may have produced a seal in the drilled area and only the pressure inside the inner container was released (reference Table 4).

Table 4

INTERNAL GAS PRESSURES

Source Number	Pressure (psia) Inner	Pressure (psia) Outer
M-75	4.7	29.8
M-259	1.5	Not detectable
M-472	16.6	Not detectable
M-493	12.5	11.7
M-619	9.5	Not detectable
M-1166	0.7	Not detectable
M-1190	27.0	Not detectable

None of the sources indicated any appreciable helium content. It is probable that most of the helium produced from plutonium-239 decay was contained in the crystal lattice of the Pu-Be (reference Table 5). (D. L. Fleming)

Metallographic Analysis of the Outer Container, Inner Container, and Fuel

Introduction Destructive post mortem analysis was performed on 10 Pu-Be neutron sources. A summary of the tests performed on the sources, including destructive analysis, is shown in Appendix B.

Table 5

MASS SPECTROMETRIC ANALYSIS OF COLLECTED GAS SAMPLES

	M-75		M-493		
Source	Inner (%)	Outer (%)	Inner (%)	Outer (%)	M-619 (%)
Argon	39.37	16.02	55.11	9.03	73.65
Hydrogen	0.19	0.00	0.13	0.17	0.04
Helium	0.00	0.00	0.12	0.00	0.11
Water	0.13	0.24	0.26	0.23	0.29
Nitrogen	48.66	72.40	35.22	89.79	20.70
Oxygen	11.60	11.31	9.12	0.71	5.21
Carbon Dioxide	0.04	0.03	0.04	0.00	0.00

(H. S. Carden)

Test Procedure Several nondestructive tests, including radiographic inspection, were made on the sources prior to destructive analysis. The purpose of such studies was to determine the integrity of the stainless steel outer container welds. Metallographic examination, x-ray microprobe analysis, and oxygen and nitrogen studies were performed during the destructive investigation. Destructive analysis was done after the gas pressure studies. Two of the sources were not subjected to the gas analysis. One of these (M-436) was found to be leaking prior to the special tests, and the other (M-1019/Schlumberger) was known to be contaminated, suggesting a severe crack or leak in the outer container.

Sectioning The sources were sectioned in a radioactive glovebox, using a circular saw with an alumina oxide cutting blade. As indicated in Figure 5, all welds, including small sections of the top, sidewall, and bottom, were taken for metallographic examination.

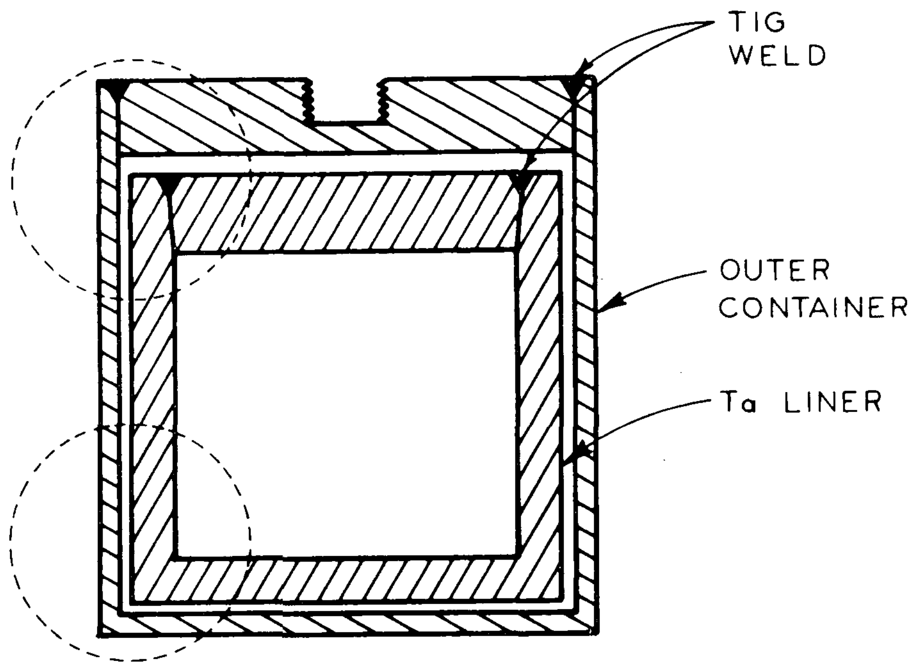

FIGURE 5 - Source areas subjected to metallographic examination (indicated by dotted circles).

Results During sectioning, various visual observations were made on each source and these are summarized in Table 6. With the exception of source M-1019 no visible cracks were noted in the outer container. Table 7 summarizes the results obtained during metallographic examination. From this table it can be seen that a few small cracks were found. The two sources, M-71 and M-1053-S, were the most severely cracked. Photomicrographs of these two sources are shown in Figures 6 and 7. Electron microprobe analysis was performed on the crack shown in Figure 6. No difference in composition was found in the region immediately adjacent to the crack and the second phase boundaries away from the crack. These areas were slightly enriched in oxygen, silicon, sulfur, and carbon with slight depletion in iron. This merely suggests a concentration of impurities which results when the lower melting constituent solidifies last. Microhardness values measured in the weld area varied between 150 and 200 Diamond Pyramid Hardness (DPH) which is typical of stainless steel. It is not expected that under normal conditions that these cracks would continue to propagate. It is also possible that the cracked areas are rather localized and are not necessarily continuous around the weld.

Table 6

VISUAL OBSERVATIONS

Source	M-259	M-1166	M-472	M-1190	M-75	M-1053S	M-9	M-1019	M-436	M-71
Outer Weld Visible Cracks	No	No	No	No	No	No	No	No	No	No
Interior of Outer Container	Traces of green oxide layer	Clean and bright	Clean and bright	Clean and bright	Clean and bright	Clean and bright	Clean	Clean	Clean	Clean and bright
Exterior of Inner Container	Brownish in color	Clean and bright	Brownish in color	Dull gray	Purplish in color	Greenish oxide layer	Dull metal color	Fairly clean	Royal blue ring mid body brownish	Clean
Inner Weld Visible Cracks	Pulled away from body	Bluish gray color around top weld	No	No	Small crack	Ruptured at weld	White residue top weld	Greenish ring around weld area	No	No
Condition of Fuel	~60% oxidized dust	Solid dull gray chunk	Dull gray chunk	Bright silver chunk	Dull gray top of fuel purple	Bright silver chunk	Dull gray on surface	~50% oxidized dust	Dull gray chunk	Bright silver
Outer Capsule Body Condition	Green oxide layer	Good	Good	Good	Brownish in color	Good	Good	Black in color	Dull metal	Good
Inner Capsule Body Condition	Brownish in color	Good	Good	Good	Good	Huge ruptured zone	Good	Fairly clean	Bluish to dark blue ring	Good

Table 7

METALLOGRAPHIC OBSERVATIONS

Source	M-259	M-1166	M-472	M-1190	M-75	M-1053S	M-9	M-1019	M-436	M-71
Top Weld Cracks	Slag inclusion	Small crack	Small crack	No	Small crack	Large crack	Slag inclusion	Completely severed	Slag inclusion	No
Bottom Weld Cracks	Slag inclusion	Slag inclusion	No	Small crack	No weld corner machined	Small crack	Slag inclusion	No weld corner machined	Slag inclusion	Small crack
Reaction Products	Yes	No	No	No	Yes	Yes	Yes	Outer edge	Yes	No
Porosity	Hole in bottom weld	No	No	No	No	No	No	Small amount	No	No
No. of Weld Passes Top Welds	4	2	1	1	1	1	3	1	2	1
No. of Weld Passes Bottom Welds	1	1	1	1	Corner machined	2	1	Corner machined	1	1
Crack Dimension	--	1 mil	4 mil	2.5 mils	--	13 mils / 5 mils	--	--	--	~5 mils

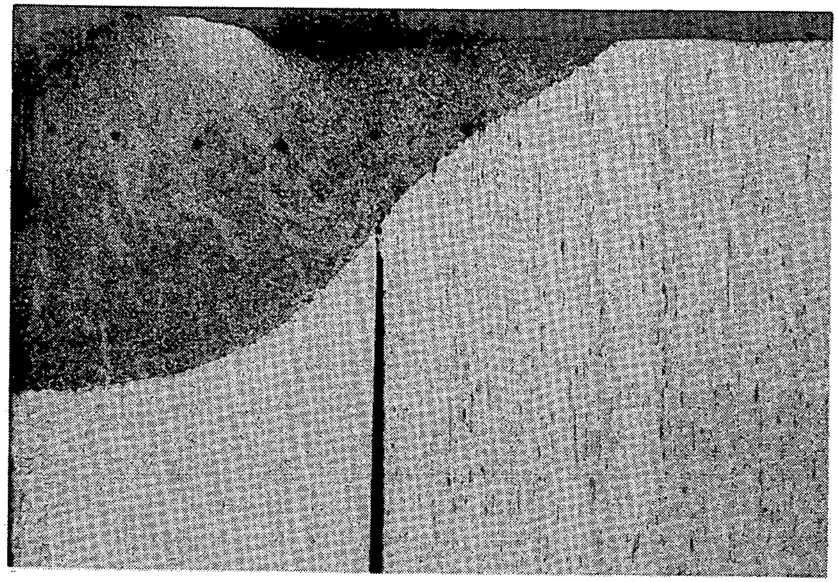

FIGURE 6 - Source M-71; stainless steel outer container; top weld (60.5X).

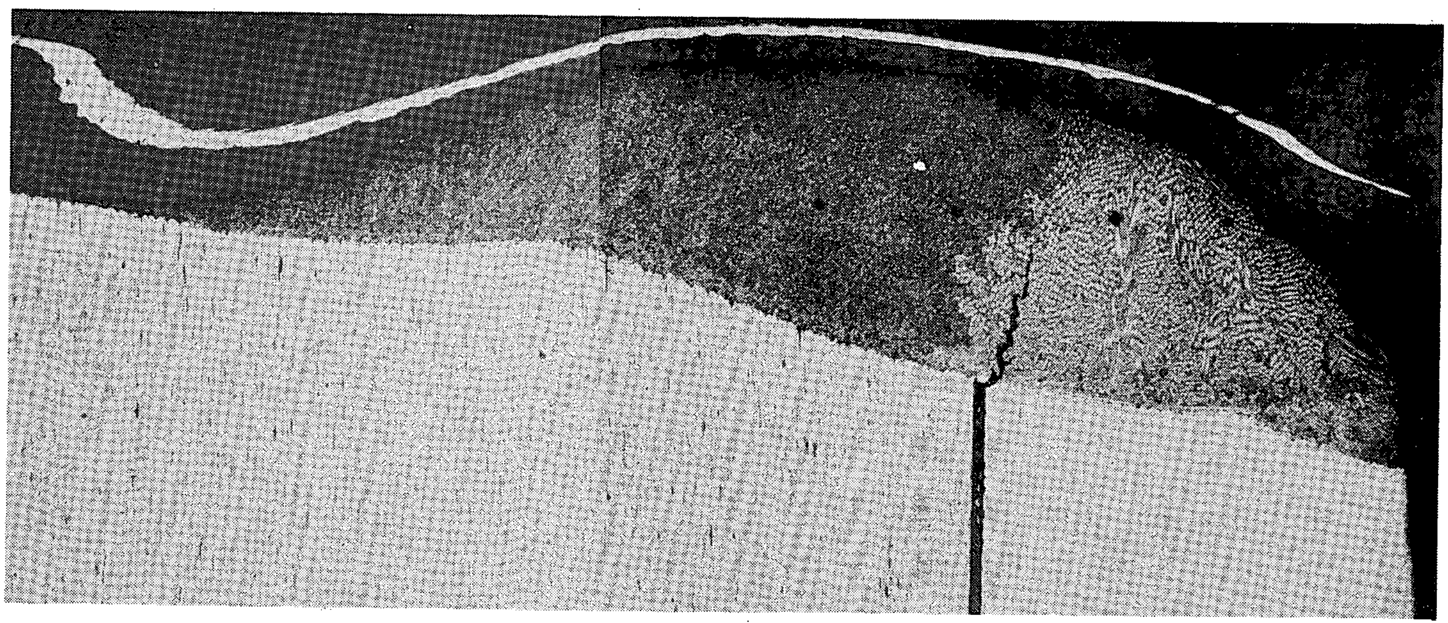

FIGURE 7 - Source M-1053S; stainless steel outer container; top weld (60.5X).

Examples of a slag inclusion in the weld joint are shown in Figures 8 - 11. The rounded ends to these separations are typical of this type of artifact. Round-ended separations such as these would not be expected to propagate. A small crack is shown in Figure 12. Under certain circumstances a sharp-ended crack could conceivably propagate, but in a ductile material such as 304 stainless steel this is not probable.

Photomicrographs of the top and bottom welds of source M-436, which had exhibited a leak, are shown in Figures 13 - 15. The small cracks or pores emanating from the side of the joint may have been responsible for the leaks. The reason for this phenomenon is not known at this time. Although corrosion could be the cause, electron microprobe analysis could not detect any reaction product in these areas. Either these are voids, or the reaction product pulled out during metallographic preparation.

Oxygen analyses were performed on the tantalum inner container of each source, and these are summarized in Table 8. Oxygen pickup probably occurred during welding.

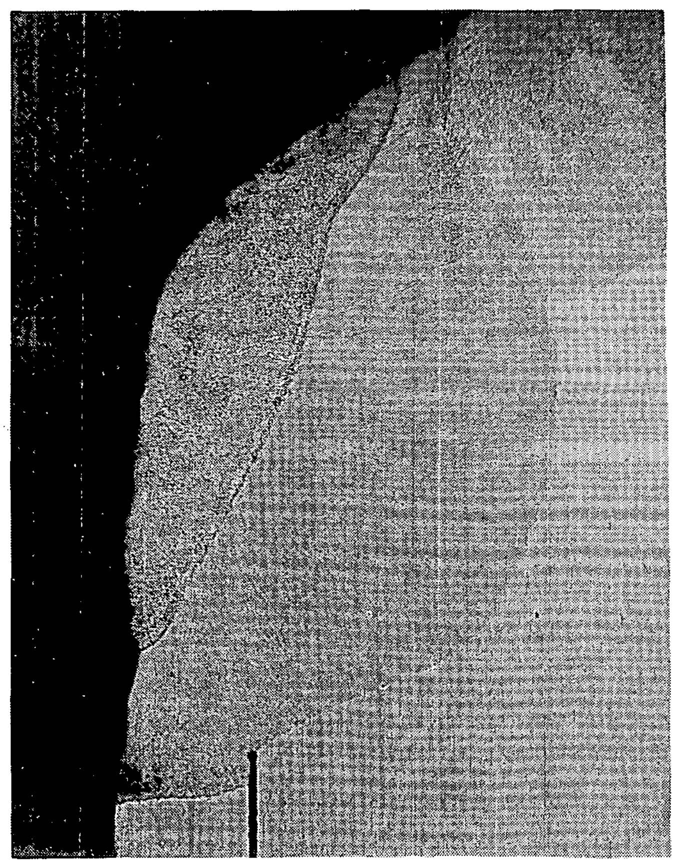

FIGURE 8 - Source M-259; stainless steel outer container; top weld (24.8X).

Weld Zone

Parent Material

FIGURE 9 - Source M-259; stainless steel outer container; top weld (620X).

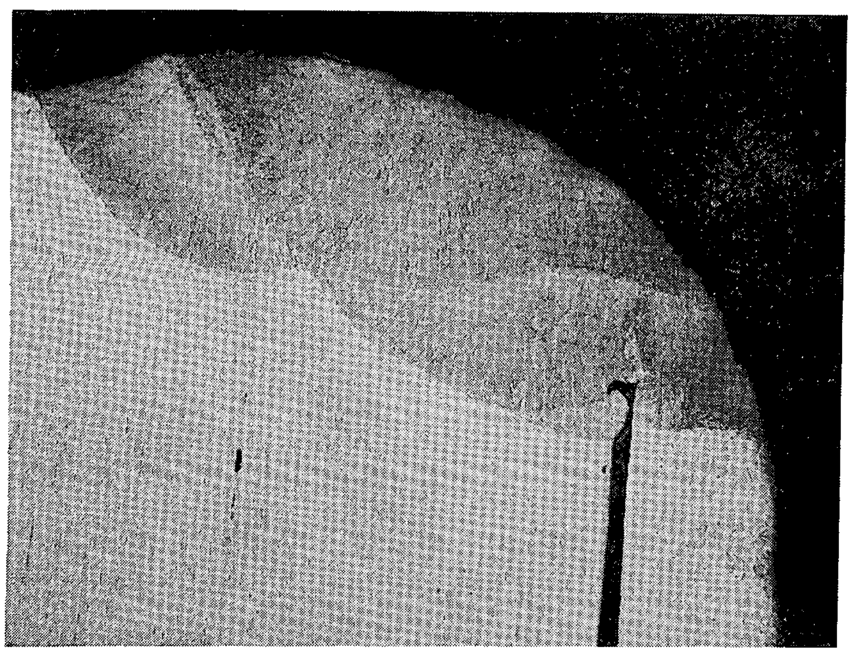

FIGURE 10 - Source M-9; stainless steel outer container; top weld (24.8X).

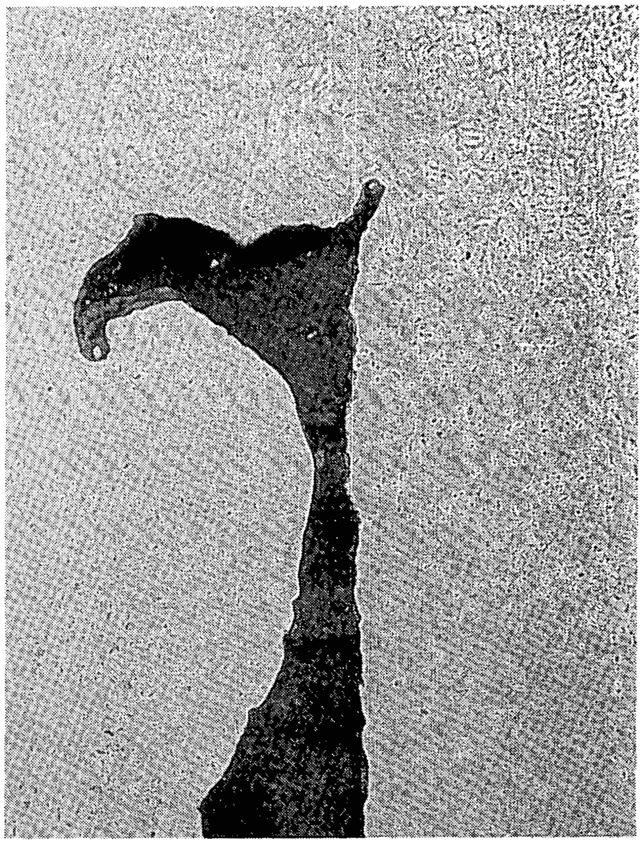

FIGURE 11 - Source M-9; stainless steel outer container; top weld (250X).

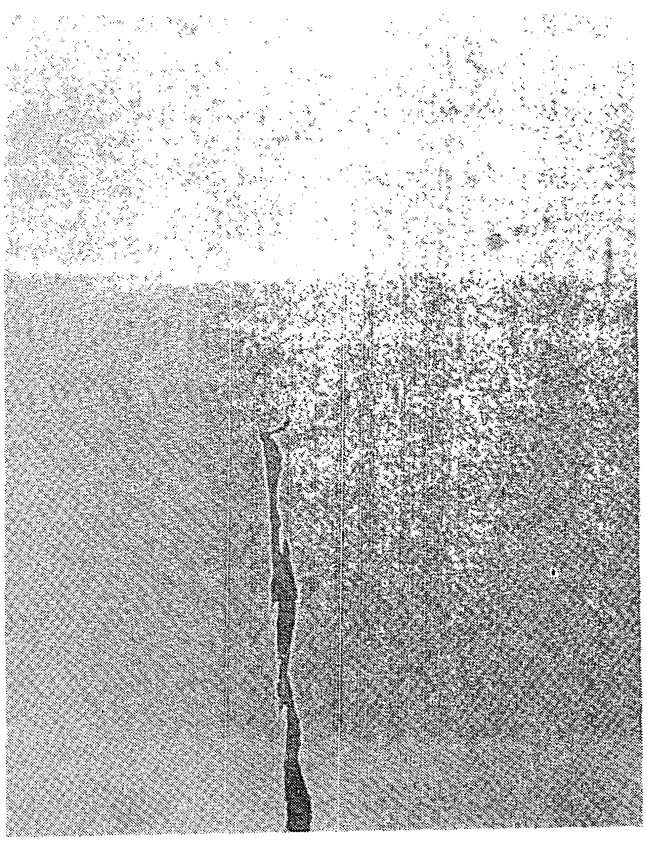

FIGURE 12 - Source M-1166; stainless steel outer container; top weld (250X).

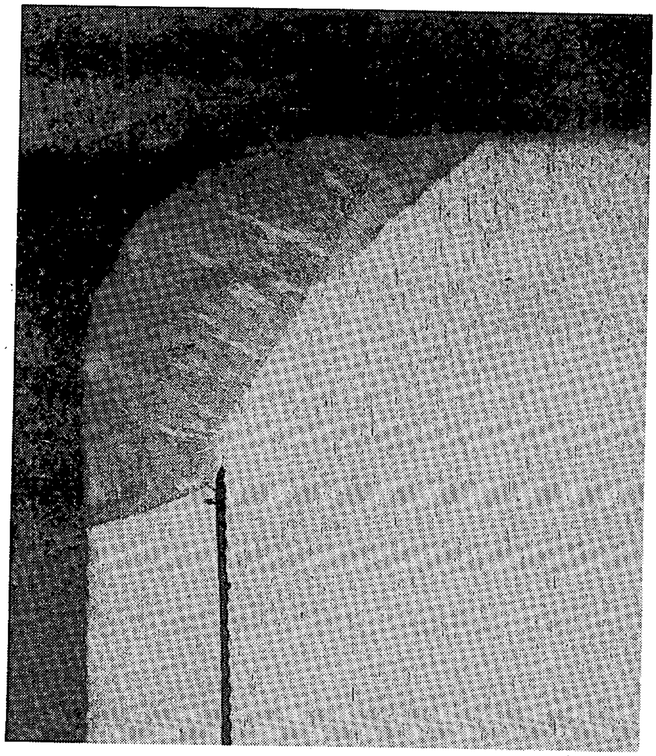

FIGURE 13 - Source M-436; stainless steel outer container; top weld (24.8X)

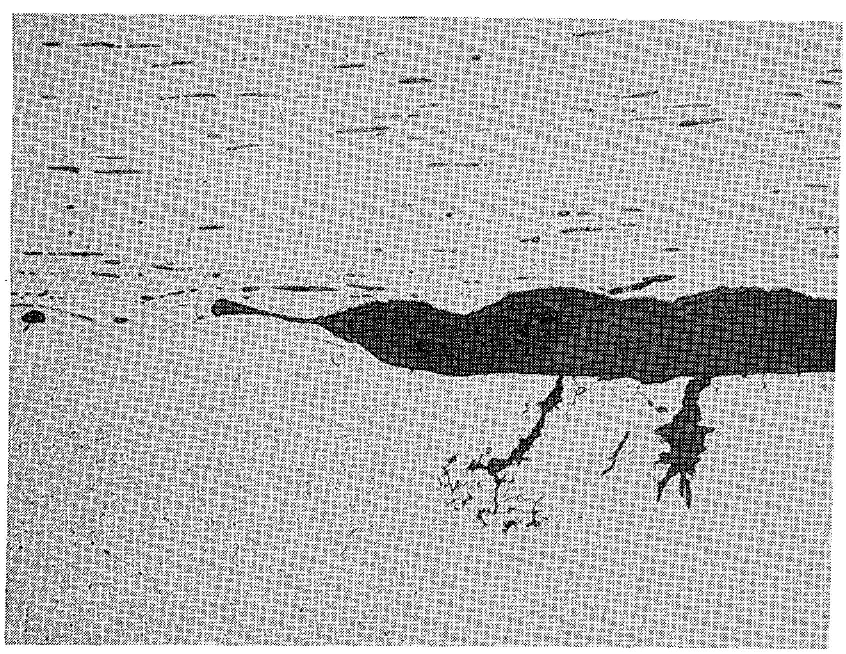

FIGURE 14 - Source M-436; stainless steel outer container; top weld (250X).

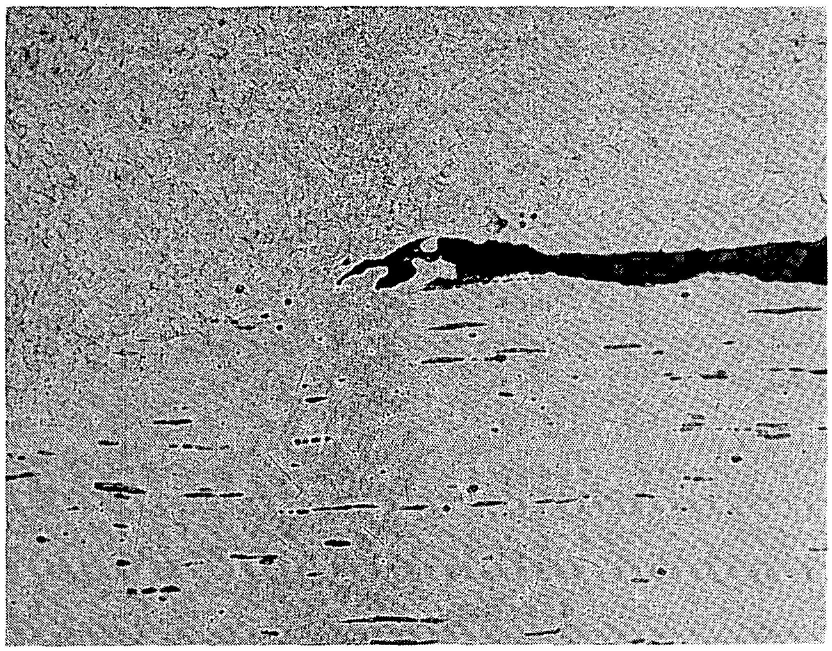

FIGURE 15 - Source M-436; stainless steel outer container; bottom weld (250X).

Table 8

SUMMARY OF OXYGEN ANALYSIS

Source M-	259	1166	472	1190	75	1053S	9	1019	436	71
Oxygen Content (ppm)	181	30	136	374	613	240	359	273	274	881[a]

[a] Analysis questionable due to wide variation in data.

Examination of M-1019 showed the weld of the outer container, a martensitic steel as shown in Figure 20, to be completely severed from the sidewall; the cap lifted off. Figure 21 shows the weld area. Some small voids are observed, which may be attributable to gas evolution during welding. As indicated in Table 7, the bottom corners of the capsule were machined from one piece, not welded. Visual observations made during sectioning are listed in Table 6.

No cracks were observed in these corners. Figure 22 shows a reaction product containing various concentrations of iron, oxygen, and chromium, with oxygen and silicon in one particle on the outside wall of the container. Penetration of this reaction product is readily observed. These data suggest that the source was subjected to elevated temperature which may have contributed to the degradation.

Vickers microhardness values for the Vega steel ranged from 550-600 DPH. These values are indicative of this type martensitic steel as shown in Figure 20.

As indicated in Table 7 some of the weld joints were found to have been subjected to multiple passes.

Samples of the fuel and the fuel-tantalum interface were examined and analyzed on the electron microprobe. Photomicrographs of the areas examined are shown in Figures 16-19. Superimposed on each photomicrograph is the direction of the beam and the areas examined. In each case where beryllium is indicated, it should be emphasized that this is implied by the data. The electron microprobe did not detect beryllium. The fact that great inhomogeneity exists in the fuel, usually associated with tantalum and probably beryllium, suggests that free unreacted plutonium may be present in the fuel, although no positive indication of unreacted plutonium was found.

All of the sources, with the exception of M-1019 (Schlumberger), had outer containers of 304 stainless steel. Source M-1019 was apparently made from an air hardening tool steel called Vega steel. This material nominally contains 0.7 wt % C, 2.0 wt % Mn, 1.0 wt % Cr, 1.35 wt % Mo. Electron microprobe analysis, while not quantitative in this case, indicated high iron with small amounts of manganese, chromium, and molybdenum, which tends to verify this composition.

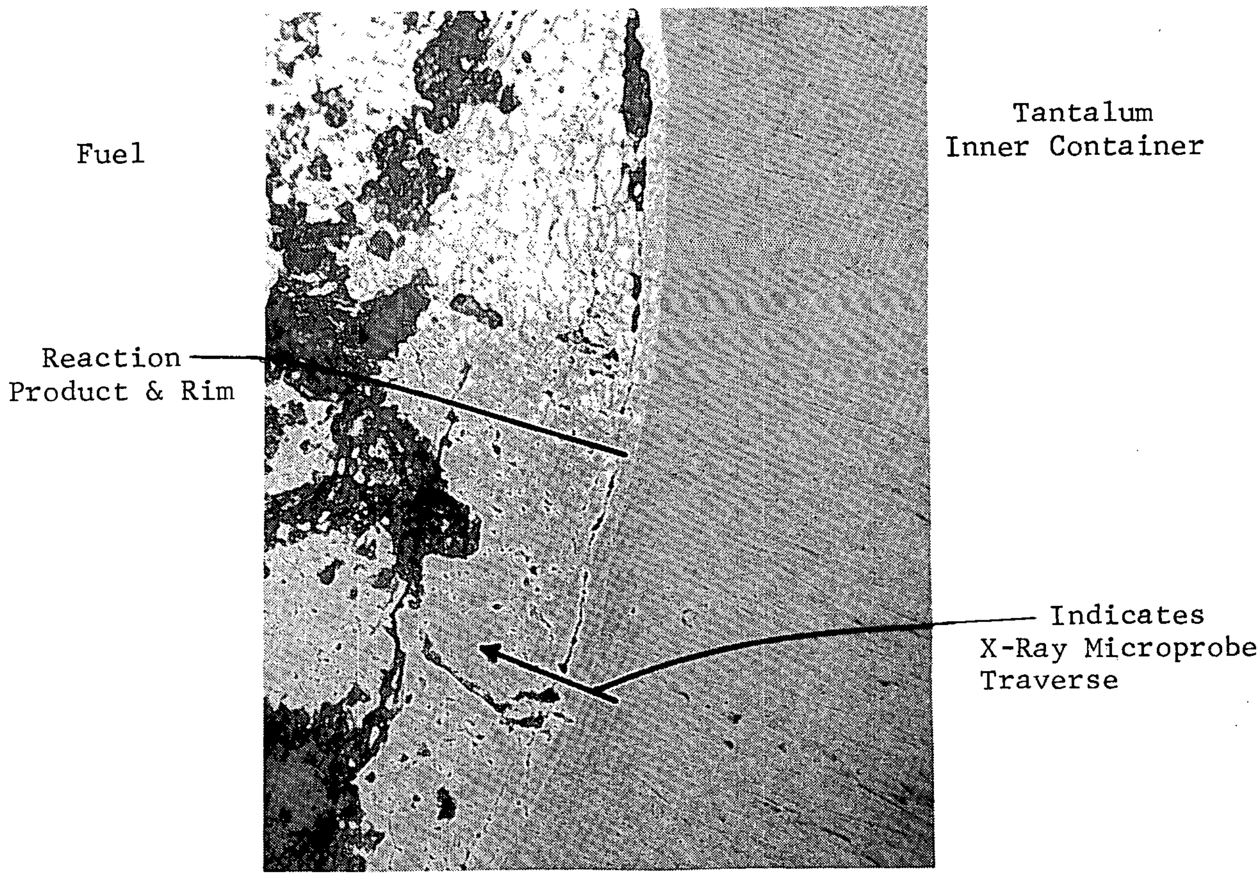

FIGURE 16 - Source M-1053S; tantalum inner container; tantalum reaction product. Reaction rim increases in plutonium, decreases in tantalum. Presence of beryllium indicated (250X).

FIGURE 17 - Source M-1053S; tantalum inner container. X-ray microprobe traverse "A": grain boundary reaction product, plutonium decrease in tantalum, oxygen may be present. Presence of beryllium indicated. X-ray microprobe traverse "B": dark gray phase, plutonium + oxygen, no tantalum (250X).

FIGURE 18 - Source M-1053S fuel chunk. X-ray microprobe traverse "A": white grain boundary reaction product, increase in tantalum, no plutonium, slight decrease in oxygen, presence of beryllium indicated. X-ray microprobe traverse "B": black phase seems to be pull-out (250X).

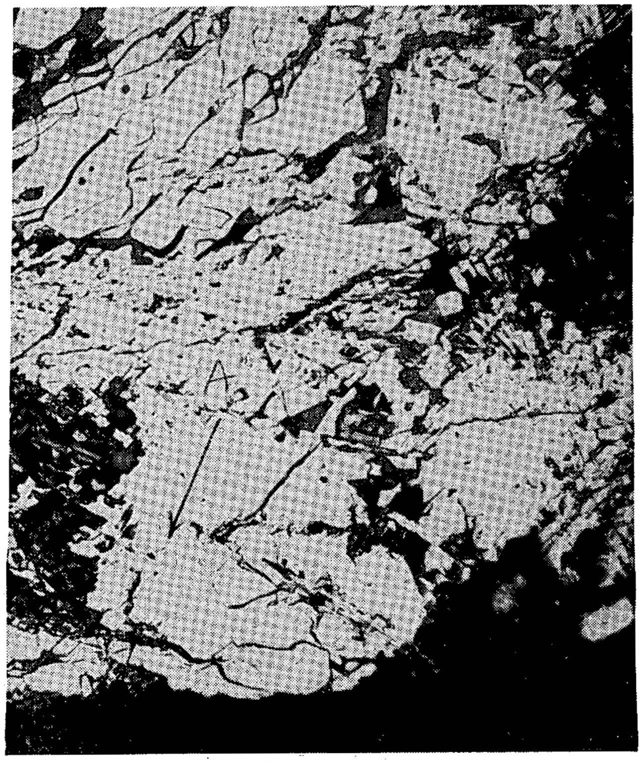

FIGURE 19 - Source M-1053S fuel chunk; x-ray microprobe traverse "A": layering, as tantalum goes up plutonium goes down, some high peaks of oxygen (250X).

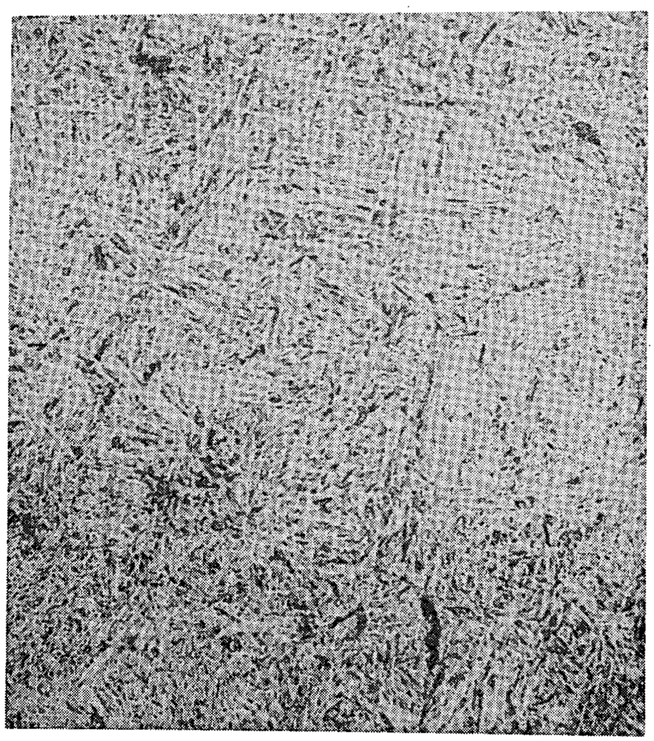

FIGURE 20 - Source M-1019; structure of Vega steel outer container (620X).

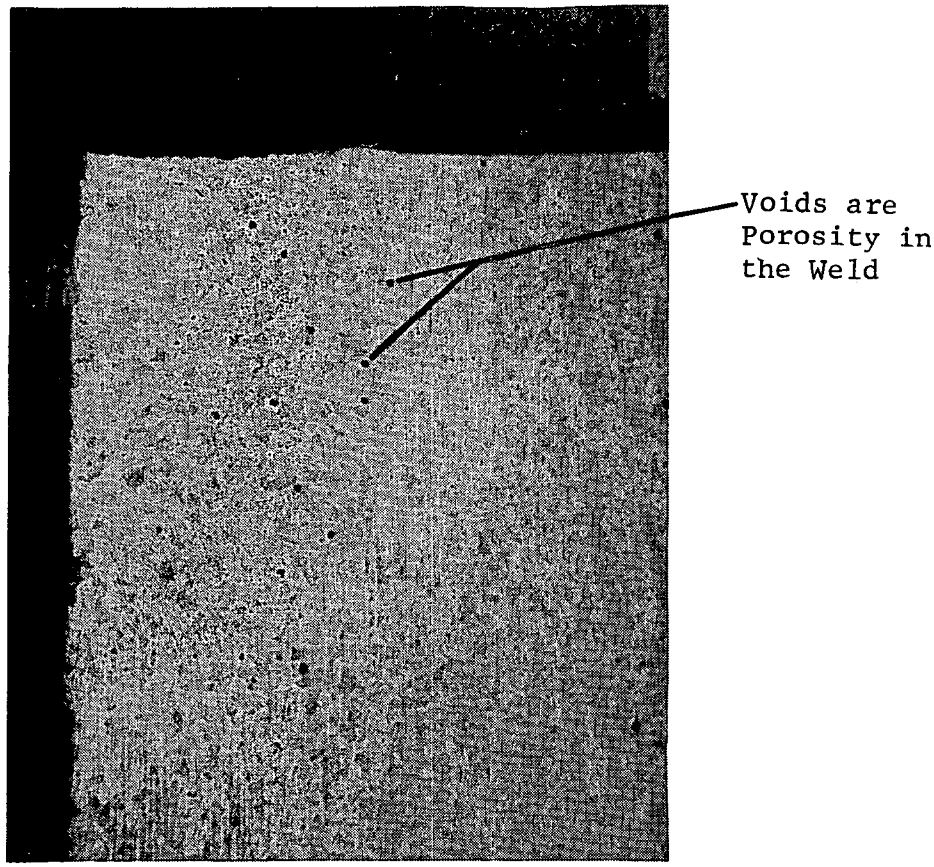

FIGURE 21 - Source M-1019; Vega steel outer container; top weld (24.8X).

FIGURE 22 - Source M-1019; Vega steel outer container; top weld outside edge (250X).

Conclusions Analysis of the stainless steel welds in nine neutron sources revealed small cracks in a total of seven welds out of 17. An additional six welds contained slag inclusions near the joint tip. Under normal conditions these cracks or inclusions would not be expected to propagate.

All of the tantalum containers contained oxygen, probably from the welding operation. In view of this and the fact that some of these welds were observed to be cracked, the tantalum should not be relied upon to contain the fuel. Composition inhomogeneities exist within the fuel which raises the possibility that unreacted plutonium exists in the sources, although neutron counting indicates a near theoretical number of neutrons for each source.

Destructive analysis results on sources M-472, 1166, and 1190 (sources which showed no significant anomalies in NDT) gave no indication of unusual or deleterious conditions. Metallography did reveal some small cracks, none of which extended completely through the weld area, and which, under normal conditions, would not be expected to propagate. (J. E. Selle, K. L. Breakall)

CONCLUSIONS AND RECOMMENDATIONS

User Survey This activity was suspended prior to completion in accordance with AEC guidance.

Nondestructive Tests (NDT) NDT techniques (radiography, leak testing; visual and dimensional checks, and alpha wipe testing) provided some significant information. Abnormalities in the condition of the fuel and inner liner were ascertained, the integrity of the outer container was verified, and significant dimensional changes were detected. The integrity of the inner container could not be assured by NDT techniques. Ultrasonics was evaluated as a method to determine weld penetration on the outer container but was found to be of no value due to irregular geometries.

Special Tests (DoT Tariff 25:173.398a) Sixteen of 18 sources which were selected without regard to inner liner integrity successfully passed the drop, percussion, heating, and immersion tests. The two which failed the immersion test had outer containers fabricated from Schlumberger-specified Vega steel.

Destructive Tests Gas pressure and gas analysis indicated that no significant pressure buildup occurred with time. Metallographic analyses indicated that the single previously reported case of source failure in use (Schlumberger) had an outer encapsulation of Carpenter Vega air-hardened tool steel.

The degree of assurance that sources having an integral outer container would pass the special accident tests was indeterminate because the presently available data represent a statistically small sample. However, the special test data reinforced the historically successful use data from a large number of sources.

General Only one source of the 1,226 Mound-fabricated sources in the field during the past decade has been involved in a reported detectable release of radioactivity (M-1019/Schlumberger). There exists a striking correlation between the use of Vega steel outer containment and failures, both in the special tests and in the field.

Recommendations If the AEC considers continued usage desirable, all Mound-fabricated sources should be subjected to the NDT evaluations consisting of radiography, leak test, visual and dimensional checks, and alpha wipe testing, with "acceptability" based primarily on the outer container material and its condition. No sources using Vega steel as the outer container material are recommended for use. Further use of "acceptable" sources presents a low level of risk regarding release of radioactivity, based primarily on historical experience and substantiated by no failures in the limited number of sources subjected to the special tests. A higher degree of assurance would be obtained by providing the sources with an outer container using current quality-controlled fabrication procedures.

APPENDIX A

NONDESTRUCTIVE TEST SUMMARY

NON-MOUND-FABRICATED SOURCES (Loan Program Inventory)

Test	No Evidence for Potential Problem	Evidence for Potential Problem	Remarks	Tested
Radiography	8			32
		2	Outer liner open	
		13	Inner liner open	
		11	Inner liner bulging	
Leak Check	12			13
		1	Leak $>3\times10^{-6}$ cc/sec	
Dose Rate	31			32
		1	High gamma for reactor activation	
Visual	31			32
		1	Deep saw cuts on side (no wipeable contamination)	
Totals	8[a]	24		32

[a] Controlled by radiography.

APPENDIX B

SOURCES SUBJECTED TO DESTRUCTIVE ANALYSIS

Source No.	^{239}Pu (g)	Mfg. Date	Recan Date	Radio-graph	Leak Check	Gas Tap	Metal-lography
M-9	16.10	11/9/56	8/10/61	x	x	x	x
M-71	15.05	10/11/57	7/13/63	x	x	x	x
M-75	15.05	10/17/57	--------	x	x	x	x
M-259	79.68	12/17/58	8/24/61	x	-	x	x
M-436	79.56	3/9/59	12/8/61	x	x	-	x
M-472	79.87	4/20/59	3/19/62	x	x	x	x
M-493	79.46	5/20/59	--------	x	x	x	-
M-619	79.92	2/8/60	7/17/63	x	-	x	-
M-1019	76.34	7/27/61	--------	x	-	-	x
M-1053-S	15.09	1/15/62	--------	x	x	x	x
M-1166	91.52		11/6/61	x	x	x	x
M-1190	15.52	3/9/62	--------	x	x	x	x

M-9 was returned from Mobil Research and Development Corporation on February 22, 1971. NDT indicated that the outer container had a minimum weld approximately 0.018 in. long, a slop-fit in the end cap, and some bulge in the walls. The inner container exhibited no indication of fuel/liner corrosion; the liner was possibly open at the weld, and a high-density material, possibly fuel, was noted between the liner and outer container in the liner weld area.

M-71 was returned from the University of California, Radiation Laboratory, after having been in a nuclear test, and was stripped of its outer container on July 13, 1963, for examination. The tantalum liner was badly cracked due apparently to temperatures reached in the test. The source was recanned in a stainless steel outer container for long-term observation. NDT indicated that the outer container was free of defects, with a minimum weld approximately 0.040 in. in length. No fuel/liner corrosion was observed in the inner liner; the weld joints were possibly open.

M-75 was returned from Armour Research Foundation of the Illinois Institute of Technology after having been subjected to spike heating. NDT indicated the outer container to be free of defects and to have a minimum weld approximately 0.025 in. long. The inner liner exhibited possible fuel/liner corrosion and a minimum weld approximately 0.025 in. long.

M-259 was returned from the USAARDC in January, 1970. NDT indicated that the outer container had a bulge in the outer wall approximately 0.008 in. long. The outer container weld was possible open (the source was not leak checked). The inner liner weld was possibly open. No alpha wipe was detected.

M-436 was returned from Gulf Logging and Perforating Company in April, 1970. NDT indicated that the outer container was free of defects. The inner container was possibly open. Upon leak checking prior to the special test series, the source was found to be leaking by the helium bubble method. No alpha wipe was detected.

M-472 was returned from Gulf Logging and Perforating Company in April, 1970. NDT indicated the outer liner to be free of defects with a minimum weld approximately 0.050 in. long. The inner liner exhibited possible fuel/liner corrosion and was possibly open. There was no indication of fuel outside the liner.

M-493 was returned from Wells Survey, Inc., for recanning, but the inner tantalum container was found to be unrepairable. It was recanned in a stainless steel outer container for purposes of local handling at Mound. NDT indicated that the outer container was free of defects, and had a minimum weld approximately 0.025 in. long. The inner liner condition noted in recanning was confirmed, with fuel possibly between the liner and outer container.

M-619 was returned from Dresser Atlas on July 15, 1971. NDT indicated the outer container to be slightly distorted. The inner liner weld appeared to be open, with the liner concaved approximately 0.012 in. near one end. No fuel/liner corrosion was noted.

M-1019 was returned from Schlumberger Well Services, Inc., in June, 1970, and reported by them to have an alpha wipe of approximately 30,000 counts/min. It was radiographed in the logging tool in which it was returned; no defects were noted in the outer container; the liner was possible open at the weld, with possible corrosion of the outer surface of the liner at the end cap.

M-1053-S was never shipped, due to a known defective liner. NDT indicated that the outer container was free of defects, and confirmed the irrepairable condition of the liner.

M-1166 was returned from Texas Nuclear on December 7, 1970. NDT indicated both the outer container and liner to be free of defects.

M-1190 was returned from John Carroll University in March, 1970. NDT indicated the outer container to be free of defects; the liner exhibited possible fuel/liner corrosion, but no apparent weld defects.