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PREPARATION OF A CESIUM (^{137}Cs)
BOROSILICATE GLASS POWER SOURCE

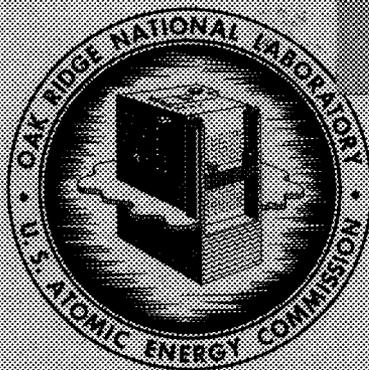
C. L. Ottinger
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ISOTOPES DEVELOPMENT CENTER

PREPARATION OF A CESIUM (^{137}Cs) BOROSILICATE GLASS
POWER SOURCE

C. L. Ottinger, E. E. Pierce, and R. W. Schaich

Isotopes Division

APRIL 1965

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PREPARATION OF A CESIUM (^{137}Cs) BOROSILICATE GLASS POWER SOURCE

C. L. Ottinger
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ABSTRACT

A cesium (^{137}Cs) borosilicate glass power source with 125 watts thermal output was prepared at the Isotopes Development Center, Oak Ridge National Laboratory (ORNL). Techniques for the preparation of the fuel material, its casting into the proper form, and the fabrication of the final source capsules were developed. This paper describes the chemical processing, fuel preparation, source encapsulation, heat output measurements, capsule leak-rate determinations, and measurements on the loaded shipping cask.

INTRODUCTION

In 1961 a program was initiated at the ORNL Isotopes Development Center to develop techniques for the preparation of a 125-watt (thermal) ^{137}Cs power source for use in an undersea application. The preferred fuel form would contain the ^{137}Cs in a glass with the highest practical power density and a low leach rate in sea water.

Fused cesium (^{137}Cs) aluminosilicate was considered as a source material since it had been shown to have low solubility in most aqueous systems and to have a negligible release of ^{137}Cs at elevated temperatures.¹ However, the ^{137}Cs concentration in cesium (^{137}Cs) aluminosilicate is too low (6-7 curies/g) for applications requiring a concentrated heat source; so the investigation of ^{137}Cs source materials was extended to the cesium glasses.

Calcination of cesium tetraphenylboron in air yields $\text{Cs}_2\text{O} \cdot \text{B}_2\text{O}_3$,² which can be reacted with SiO_2 at high temperature to produce cesium borosilicate glass. All of the operations can be carried out by remote manipulator cell operations. For this reason cesium (^{137}Cs) borosilicate glass was selected

as the fuel form for this project, and the development of a manufacturing process for this material was undertaken.

CHEMICAL PROCESSING

Prior to full-scale processing at the Fission Products Development Laboratory, a series of experimental cesium glass preparations was made. These included the preparation of inactive cesium glasses traced with ^{137}Cs , with cesium contents ranging from 30-55 wt %, and the preparation of small amounts of fully active cesium (^{137}Cs) borosilicate glass in a hot cell. On the basis of the results of these experimental runs, a flow sheet (Fig. 1) was developed to produce cesium (^{137}Cs) borosilicate glass with a cesium content of 45-50 wt %. It will be recalled that about one-third of the atoms in fission-product cesium are ^{137}Cs . The procedure was as follows.

1. Product-grade fission-product $^{137}\text{CsCl}$ (33 curies of ^{137}Cs per gram of cesium) was dissolved in 0.1 M acetic acid.
2. A predetermined amount of 140-mesh SiO_2 was added to the $^{137}\text{CsCl}$ solution.
3. The slurry was heated to 70°C and sodium tetraphenylboron, at a ratio of 3.2 grams per gram of cesium, was added.
4. The system was cooled to 30°C ; then the slurry was filtered through a medium porosity alundum filter thimble.
5. The precipitate, in the alundum thimble, was calcined in air at 450°C for one hour then at 700°C for one hour.
6. The calcined precipitate was blended in a Waring Blendor; then it was assayed for ^{137}Cs content by calorimetry.

Nine precipitations were necessary in order to yield sufficient material to complete the source. The major operational problem that was encountered during the first few runs was the tendency of the cesium tetraphenylboron precipitate to stick to the walls of the precipitator. Since the precipitate could not be washed off the precipitator walls by normal water spray flushing, the quantities of chemicals added had to be varied from batch to batch in order to maintain the desired ^{137}Cs content in the final glass. Adjustment of the agitation and washing cycles resulted in an operating procedure which was used successfully on the last four runs. The

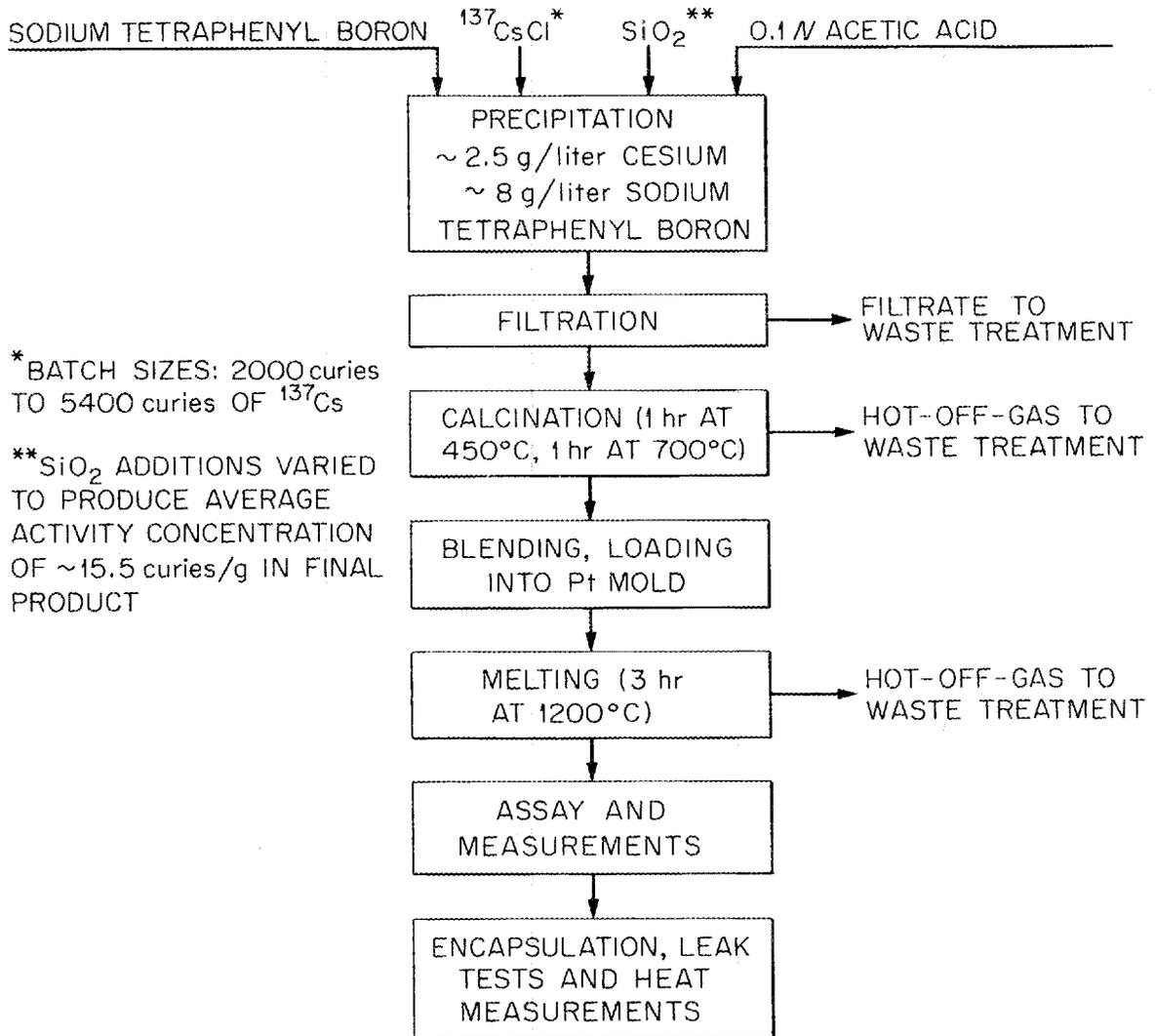


Fig. 1. Cesium-137 Borosilicate Glass Project - Flow Diagram

yield was 96%, as shown in Table 1, with most of the loss in equipment holdup.

Table 1. Cesium (^{137}Cs) borosilicate precipitations

| Batch number | Starting ^{137}Cs analysis, curies | Calcined precipitate | | Total curies |
|--------------|---|----------------------|----------------------------------|--------------|
| | | Weight, g | Activity concentration, curies/g | |
| 1 | 4,940 | 315.0 | 13.5 | 4,250 |
| 2 | 2,760 | 165.0 | 15.0 | 2,470 |
| 3 | 2,050 | 131.0 | 15.0 | 1,960 |
| 4 | 4,200 | 266.5 | 15.0 | 3,980 |
| 5 | 4,500 | 285.2 | 15.0 | 4,270 |
| 6 | 2,000 | 126.8 | 17.0 | 2,160 |
| 7 | 4,700 | 276.0 | 17.0 | 4,680 |
| 8 | 5,200 | 305.0 | 17.0 | 5,090 |
| 9 | 5,400 | 322.0 | 17.0 | 5,480 |
| Totals | 35,750 | 2,192.5 | | 34,340 |

SOURCE FABRICATION

Pelletization

Six cesium (^{137}Cs) borosilicate composites were prepared by blending selected batches of the calcined powder in a Waring Blendor. The composites were loaded into platinum cups; then they were melted individually in small furnaces at 1200°C for 3 hr.

The six melts were weighed and measured, and the excess platinum was removed by trimming with a sharp knife. The total ^{137}Cs loss by volatility and mechanical losses (calculated from weight-loss measurements) was approximately 2,500 curies (~10%). Heat output measurements were run on the individual melts using calorimetric techniques developed by Posey et al.⁵ The data pertaining to these glass melts are listed in Table 2. Since the density values are based on maximum dimensions, they are lower than the true physical density of the glass. The range of power density, from 0.19 watts/ml to 0.24 watts/ml, is probably representative for a series of cesium (^{137}Cs) borosilicate glasses made by the technique described.

Table 2. Data on cesium (^{137}Cs) borosilicate glass melts

| Melt number | Weight, g | Density, g/ml | Activity concentration, curies/g | Heat output, watts $^{137}\text{Cs}^*$ | Power density, watts/ml | Total curies |
|-------------|--------------|---------------|----------------------------------|--|-------------------------|--------------|
| 1 | 232.0 | 3.06 | 13.7 | 14.41 | 0.19 | 3,200 |
| 2 | 316.0 | 3.24 | 14.8 | 20.58 | 0.21 | 4,670 |
| 3 | 316.4 | 3.16 | 14.8 | 20.61 | 0.21 | 4,690 |
| 4 | 308.5 | 3.05 | 16.1 | 24.45 | 0.24 | 4,970 |
| 5 | 281.2 | 3.02 | 16.1 | 22.37 | 0.24 | 4,550 |
| 6 | <u>286.8</u> | 3.00 | 16.1 | <u>22.68</u> | 0.24 | <u>4,600</u> |
| | 1,740.9 | | | 125.10 | | 26,680 |

* Corrected for ^{134}Cs contribution (2.8%).

Encapsulation

The six completed cesium (^{137}Cs) borosilicate glass melts (Fig. 2) were loaded into two Hastelloy inner capsules. The inner capsules were welded remotely (Fig. 3) under argon containing ^{85}Kr tracer. Each inner capsule was cleaned and leak-tested for ^{85}Kr . The inner capsules were inserted into Hastelloy outer capsules which were then welded remotely under argon with ^{85}Kr tracer. The completed capsules (Fig. 4) were leak-tested for ^{85}Kr , then cleaned to an average smear count of <200 d/min for a single smear over the entire surface. All of the leak rates, determined by the ^{85}Kr tracer method, were below the limit of detection of 0.44 ml/year (see Appendix).

Assembly and Measurements

Calorimetric measurements on the completed capsules showed 55.6 watts for No. 1 and 69.5 watts for No. 2. Capsule No. 1 was placed in the bottom of the cavity of a 2.0-in.-thick depleted uranium shield (Fig. 5), and capsule No. 2 was placed on top of capsule No. 1. The free space above the capsules was approximately 1/8 in.

Three thermocouples were attached to the shield--one on the bottom and two on opposite sides of the cylindrical portion. As shown in Table 3, the shield surface temperature was fairly uniform, with the temperature on the bottom surface running slightly less than that on the sides.

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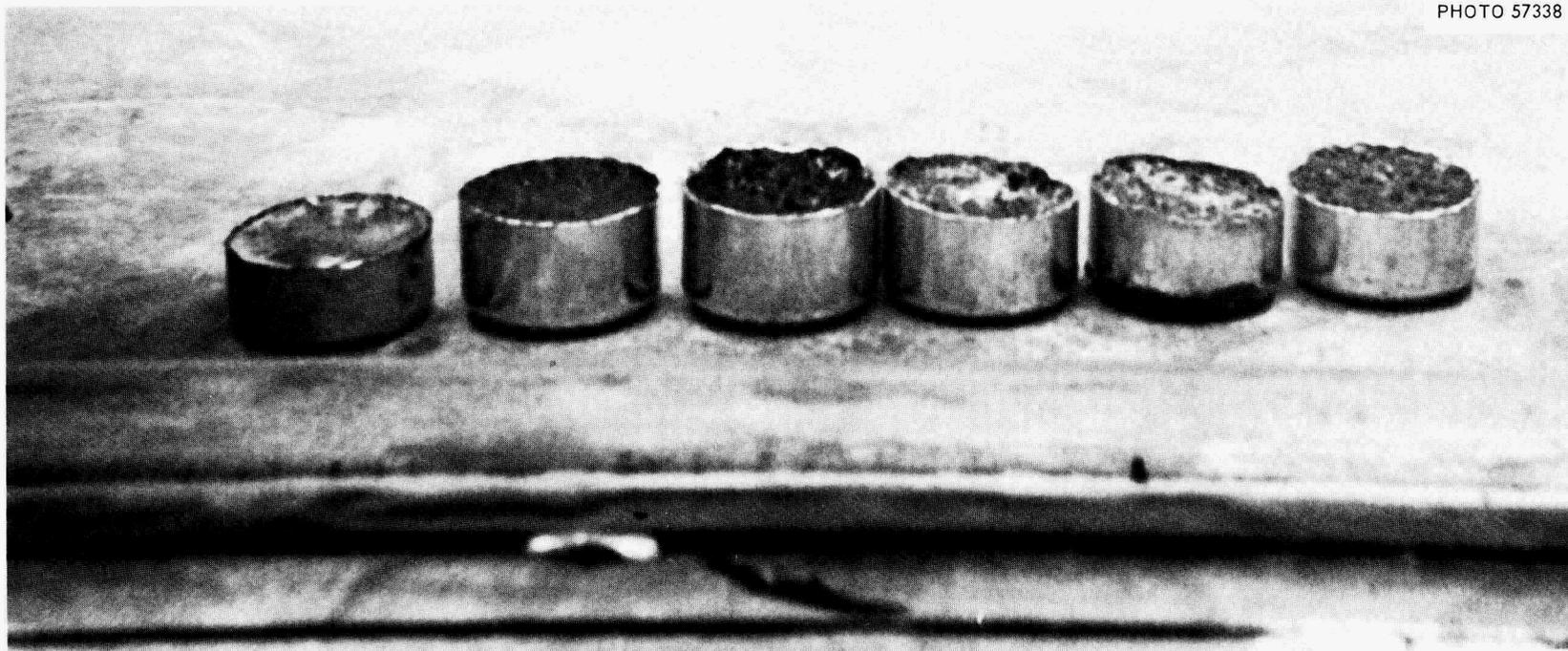


Fig. 2. Cesium (^{137}Cs) Borosilicate Glass Melts

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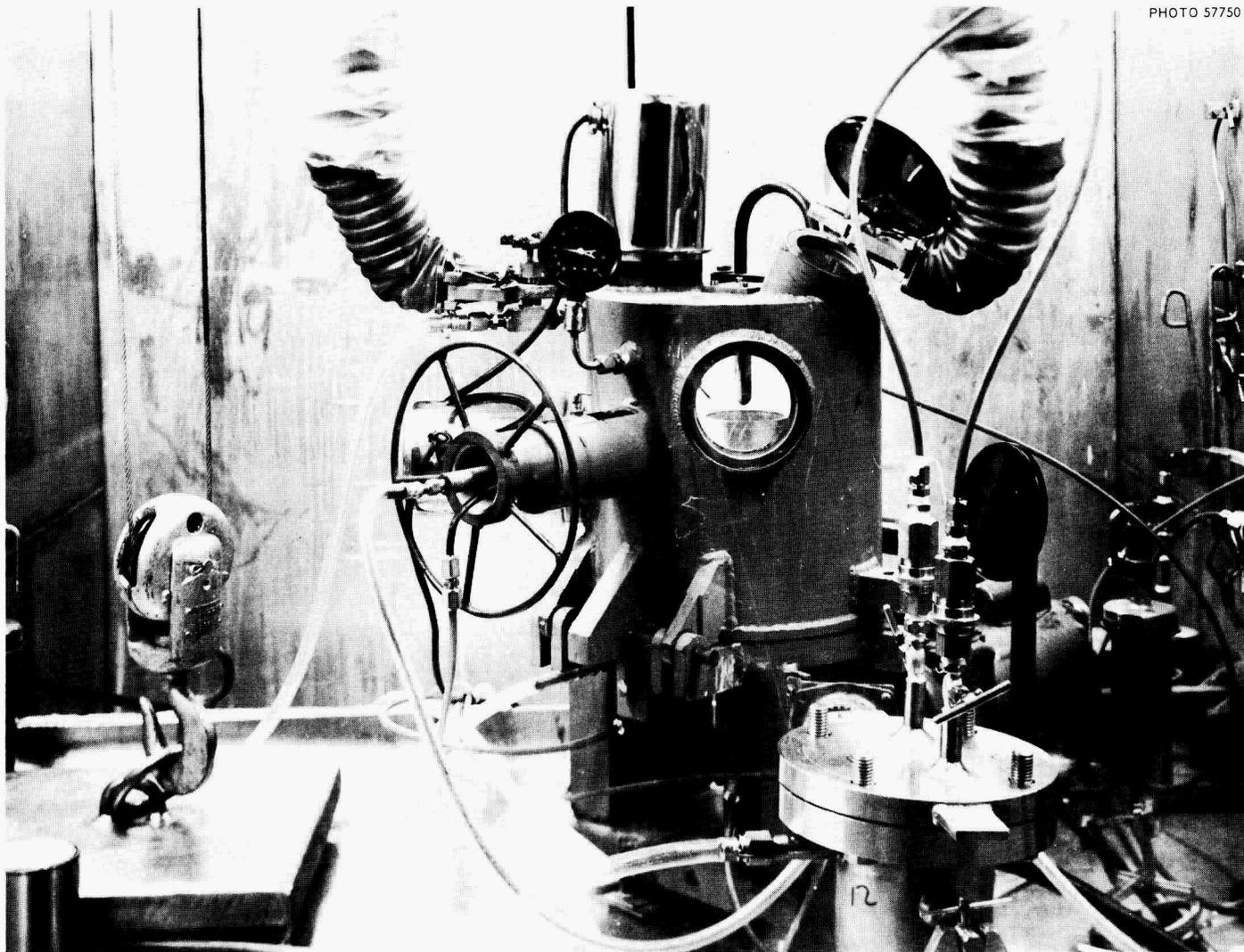


Fig. 3. Remote Pressure Welder

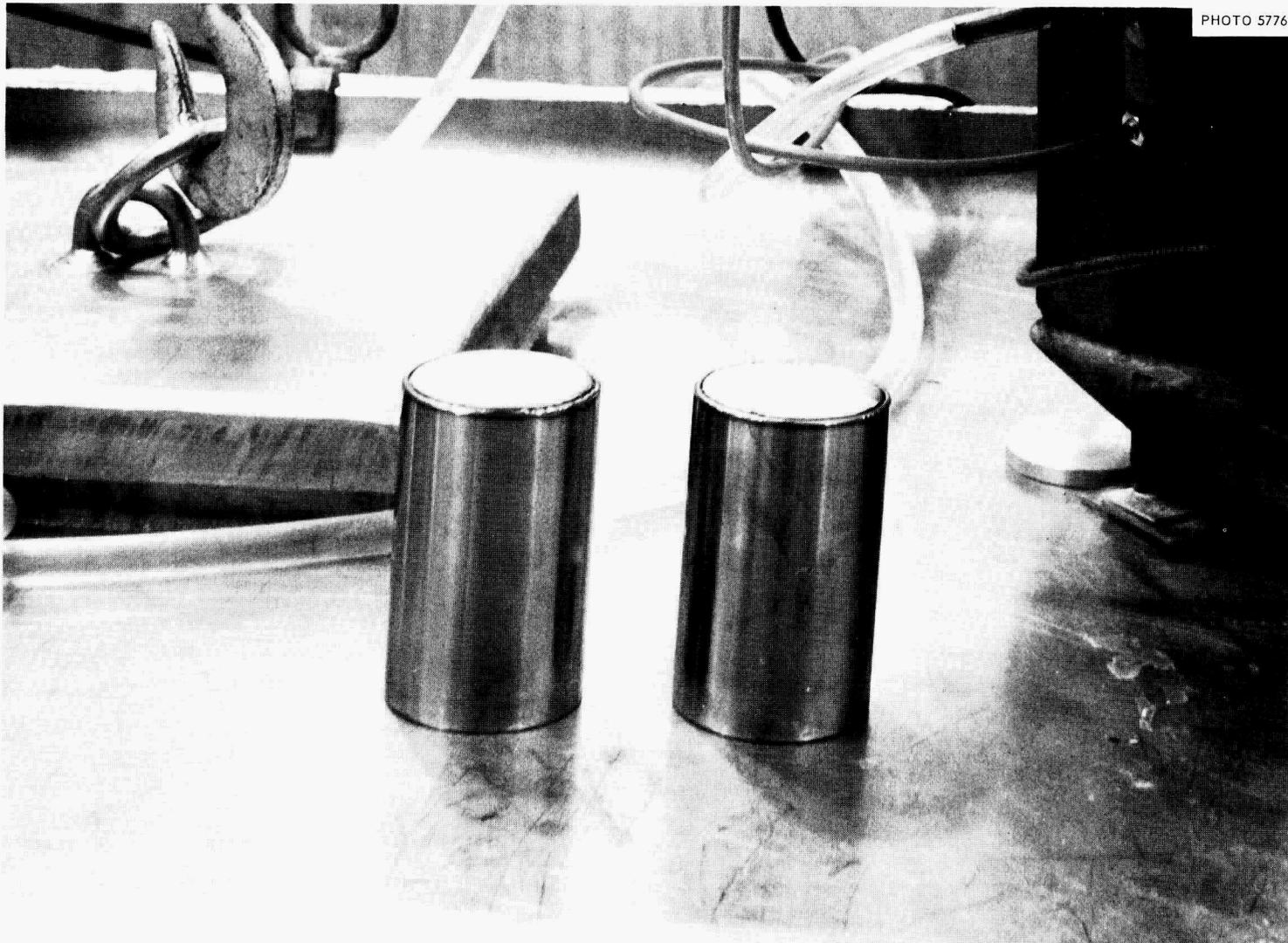


Fig. 4. Cesium (^{137}Cs) Borosilicate Fuel Capsules

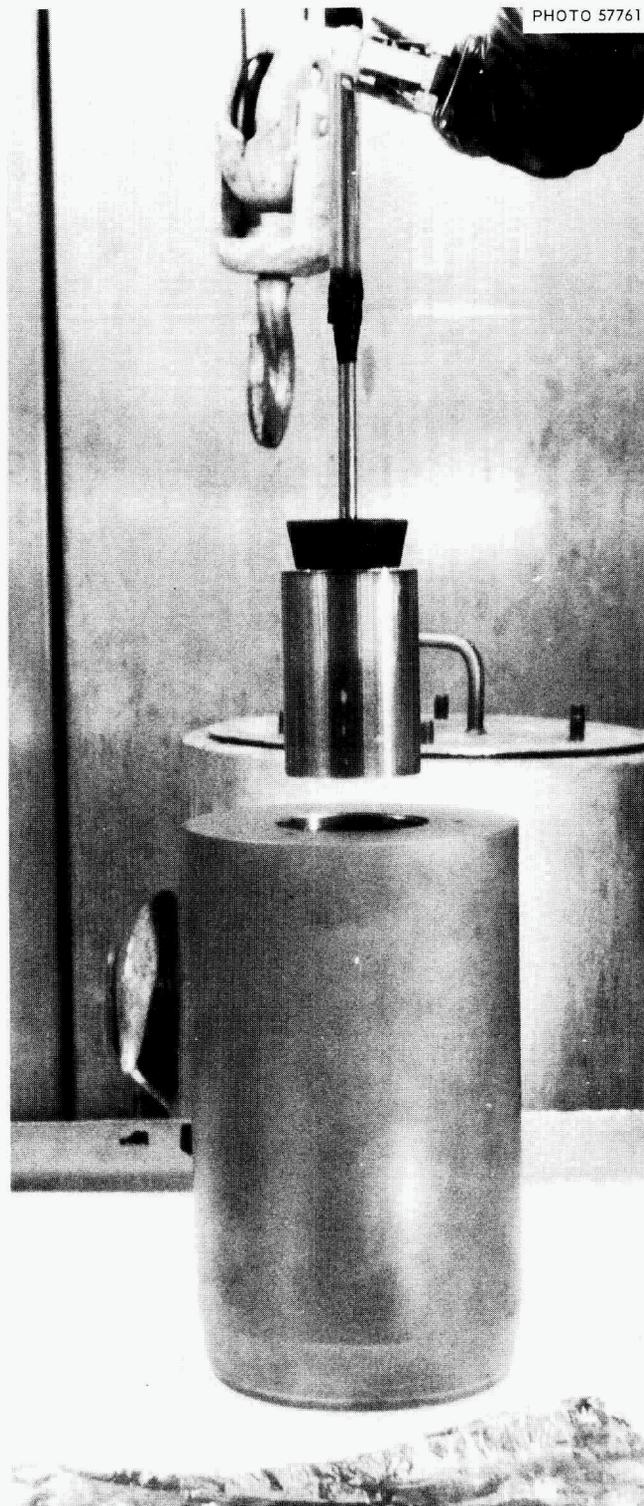


Fig. 5. Fuel Loading of Cesium (^{137}Cs) Borosilicate Power Source

Table 3. Cesium (^{137}Cs) borosilicate power source temperature measurements

| Time, min | May 28, 1962 | | | Time, min | May 29, 1962* | | |
|--------------|--------------|------|------|--------------|---------------|------|------|
| | Temp, °C** | | | | Temp, °C** | | |
| | Bottom | Side | Side | | Bottom | Side | Side |
| 0 | 80 | 88 | 83 | 0 | 72 | 80 | 80 |
| 15 | 81 | 85 | 85 | 15 | 71 | 81 | 81 |
| 30 | 83 | 85 | 85 | 30 | 71 | 81 | 83 |
| 45 | 83 | 86 | 85 | 45 | 80 | 82 | 83 |
| 60 | 83 | 86 | 86 | 60 | 80 | 82 | 84 |

*These readings were taken with the shield resting on wooden blocks.

**Ambient temperature was 30°C in all cases.

The uranium shield was placed in the shipping cask (Fig. 6) and allowed to remain overnight to measure the temperature under transport conditions. Table 4 gives the temperatures thus measured; they showed that no temperature problems should be expected during transport.

Table 4. Cesium (^{137}Cs) borosilicate power source shipping cask temperatures

| Time, hr | Temp, °C | | |
|-------------|----------|----------------------------|-----------------|
| | Ambient | Between cask and shield | Cask surface |
| 0 | 30 | 72 | 30 |
| 16.25 | 27 | 72 | 41 |
| 17.5 | 27 | 72 | 41 |
| 18.25 | 28 | 71 | 41 |

The uranium shield was removed from the shipping cask and placed on its side in two wooden "V" blocks. Radiation measurements were taken with the shield in this attitude. The results of these measurements and the calculated⁴ values are shown in Table 5. The agreement between the calculated and observed readings was satisfactory for a source of this size and shape.

Smears for surface contamination were made on the uranium shield, the inside of the shipping cask, and the surface of the shipping cask just prior to loading for shipment. In all cases the contamination level was <20 d/min alpha and <500 d/min beta-gamma.

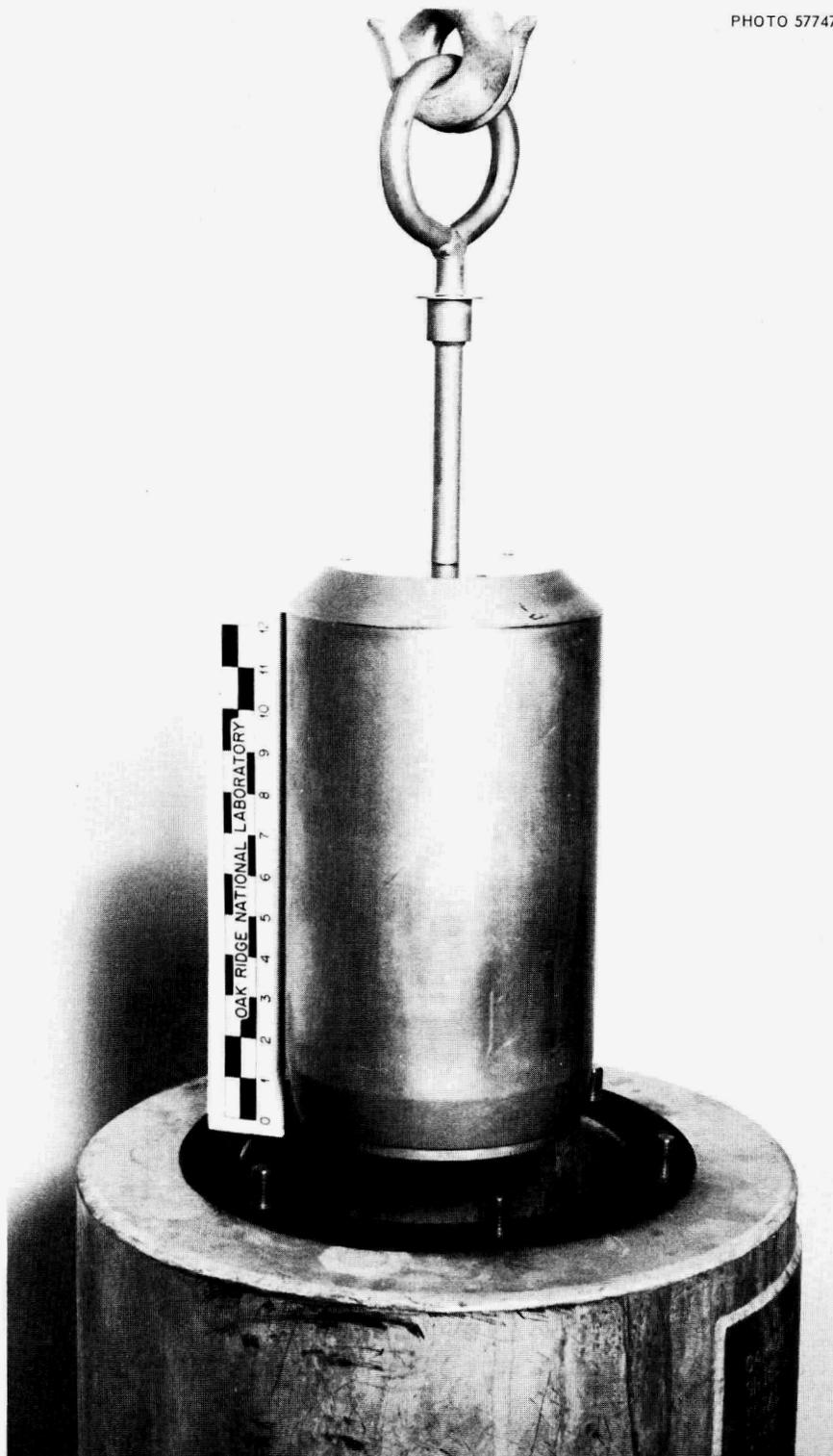


Fig. 6. Uranium Shield and Shipping Cask for Cesium (^{137}Cs) Borosilicate Power Source

Table 5. Cesium (^{137}Cs) borosilicate power source
radiation measurements

| Distance from shield, cm | Radiation (side), mr/hr | | Radiation (end), mr/hr | |
|-----------------------------|-------------------------|----------|------------------------|----------|
| | Calculated | Observed | Calculated | Observed |
| 10 | 1,030 | 1,300 | 1,520 | 700 |
| 30 | 540 | 430 | 320 | 200 |
| 100 | 190 | 66 | 38 | 34 |

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3. J. C. Posey, T. A. Butler, and P. S. Baker, Hot cell calorimetry for routine determination of thermal power generated by kilocurie sources, Proceedings of the Tenth Conference on Hot Laboratories and Equipment, Washington, D. C., 1962, pp. 263-268, American Nuclear Society.
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APPENDIX

⁸⁵Kr Leak-test Procedure for Cesium (¹³⁷Cs) Borosilicate
Fuel Capsules

All of the source capsules were welded in a sealed welder under argon. The following procedure was used.

1. The capsule, with a press-fit cap, was inserted into the welder, and the welder was sealed. The capsule cap was scored to permit transfer of atmosphere between the welder and the capsule.
2. The welder was evacuated and refilled six times with dry argon. Then it was pressurized to three atmospheres with argon containing two curies of ⁸⁵Kr.
3. The capsule was welded.
4. The capsule was removed from the welder, and the weld was inspected visually. The surface of the capsule was cleaned.
5. The capsule was inserted into the leak detector (a sealed chamber having valved lines to vacuum and argon supply and connected through a valve to an evacuated counting chamber).
6. The leak detector was evacuated and refilled with dry argon four times. Then it was filled with argon at atmospheric pressure.
7. The capsule was allowed to remain in the leak detector for four hours. The mixed Ar-⁸⁵Kr in the capsule could thus pass through any leaks in the capsule and mix with the argon in the leak detector.
8. The valve between the leak detector and the evacuated counting chamber was opened, and the system was allowed to stabilize. The counting chamber was then sealed, removed from the system, and the ⁸⁵Kr was counted.
9. The following calculations were made to determine leak rate:

| | |
|--|---|
| Free volume of welder | 12.2 liters |
| Free volume of leak detector | 1.08 liters |
| Volume of counting chamber | 1.07 liters |
| Pressure of welding gas (Ar + ⁸⁵ Kr) | 3 atm |
| ⁸⁵ Kr content | 2000 mc |
| Efficiency of counter | 1%, background count = 1000 ± 50 c/min |

a) Volume of welding gas at 1 atm = 36,600 ml

b) Concentration of ^{85}Kr in welding gas =

$$\frac{2,000 \text{ mc}}{36,600 \text{ ml}} = 0.055 \text{ mc/ml}$$

c) Disintegration rate = $0.055 \text{ mc/ml} \times 2.22 \times 10^9$
 $\text{d/min}\cdot\text{mc} = 12.2 \times 10^7 \text{ d/min}\cdot\text{ml}$

d) Count rate at 1% efficiency = $12.1 \times 10^5 \text{ c/min}\cdot\text{ml}$

e) Therefore, $1 \text{ c/min} = 0.83 \times 10^{-6} \text{ ml}$

f) Since only 50% of the leak detector gas is sampled, $1 \text{ c/min} = 1.66 \times 10^{-6} \text{ ml}$ of welding gas

g) Since the background count can vary by 120 c/min from high to low, any count rate of 120 c/min (equivalent to $2.0 \times 10^{-4} \text{ ml}$ of gas) represents an uncertain leak. For a 4-hr accumulation, this count rate is equivalent to a leak rate of 0.44 ml/year, which is the lowest leak rate detectable under the conditions described. The limit of detection may be reduced by increasing the accumulation time.

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