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H. R. Canale
Chief, Declassification Branch

CHEMICAL RESEARCH-RADIOCHEMISTRY

Section II, Group 3 (Wm. Robinson)

AEC RESEARCH AND DEVELOPMENT

SEPARATION OF FISSION GASES FROM SOLUTIONS OF URANIUM AND PLUTONIUM
Problem Assignment Number 283 MLC 3303

H. H. Adams and H. Finkelstein

With an Addendum: Interpretation of and Inferences from These Experiments
Wm. Robinson

September 4, 1945

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Sweeping of Fission Gases from Solutions of U and Plutonium

R. M. Adams and H. Finkelstein

Two experiments have been performed to compare the amounts of fission gases swept out of UH₃ solution with those amounts swept out of Pu solution under identical conditions. The charged wire technique (references given in CC-2310, p. 55) was used. The essential data are given in Table II and the results in Table III.

The procedure in the experiments was as follows: The apparatus was assembled as shown in Figs. 1a and 1b. As soon as the solution was added to the chamber and the chamber attached to the aluminum tube the flow of H₂ was started. The gauge used to measure the pressure on the graphite disc had a range of 0-60 ounces graduated in $\frac{1}{2}$ ounces and could easily be read to $\frac{1}{4}$ of an ounce. The rotameter, in conjunction with the pressure gauge, had previously been calibrated against the wet-test meter and served as a quick means of determining the flow rate at any time during the course of the experiments. The humidifier was a four foot long horizontal glass tube 40 mm in diameter fitted on both ends with rubber stoppers through which passed glass lead tubes about $\frac{1}{4}$ inch from the circumference. The humidifier was filled with distilled water to just below the level of the inlet and outlet tubes and the gas was thus humidified by passing over a four foot length of water. The nickel collecting wire was charged negatively to 1000 volts. When everything had been checked the apparatus was lowered into the thimble at CP-3 for a one-hour bombardment at 300 KW. When the bombardment was finished the apparatus was allowed to cool for about two hours before it was removed from the thimble. The nickel collecting wire was removed and dissolved in nitric acid. The chamber was detached from the aluminum tube, the solution poured out and the chamber rinsed with nitric acid which was added to the solution. Analyses were then

performed on these solutions according to the standard analytical procedures (Ba, Sr - MUC-NS-155; Zr, CL - CDC #4; Mo, MUC-WR-340 modified; Ce, CL - CDC #4 modified; Y - unpublished procedure)

Table I

Comparison of Conditions in the U and Pu Experiments

| | U Experiment | Pu Experiment |
|--------------------------|----------------------|----------------------------|
| Volume of Solution | 13.0 c.c. | 13.0 c.c. |
| Weight of U in Solution | 1.14 gm (ordinary U) | 1.0 gm (3500:1 depleted U) |
| Weight of Pu in Solution | - | 5 mg |
| Average flow rate | 338 cc/min. | 388 cc/min. |
| Gauge pressure | 25 ozs. | 30 ozs. |
| Rotameter reading | 77 | 75 |
| Pile power | 300 KW | 300 KW |
| Bombardment time | 1 hour | 1 hour |

In the plutonium experiment the depleted uranium was used to give approximately the same uranium concentration as in the uranium experiment, thus making the viscosity and density of the two solutions approximately the same.

The same apparatus was used in both experiments with the exception of the solution chamber. For the plutonium experiment an entirely new chamber was made to the exact dimensions of that used in the uranium experiment, and the graphite disc was cut from the same bar of graphite. The chambers were thus as nearly identical as possible. The percentage of various fission chains with gaseous members swept out of solution under these identical conditions are given in Table III.

Several corrections must be made on the results. The activity on the wire due to spray must be subtracted. In the uranium experiment the

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amount of spray was determined by spectrographic uranium analysis of the wire solution and found to be $10^{-4}\%$ which is negligible. In the plutonium experiment however the spray was $2.5 \times 10^{-2}\%$, as determined by α - counting, which is not negligible for those chains which are swept out to a small extent. Moreover, in the plutonium experiment 4% of any activity is due to the uranium in the 3500:1 depleted uranium oxide, which means that 4% of the total amount (i.e., the amount on the wire plus the amount remaining in the solution) of any given activity must be subtracted from the total amount found, and if the chain is swept out to a large extent then the 4% must be split up according to the distribution as determined by the uranium experiment. Also, in the case of the 86 min Ba, (parent gas, Xe^{139} with $T_{1/2} = 41$ sec) and 55d Sr, (parent gas, Kr^{89} with $T_{1/2} = 155$ sec) a correction must be made for that amount of activity which escaped deposition on the wire by virtue of the long half-lives of the gases since a gas atom spent on the average only 237 sec. in contact with the wire in the uranium experiment and 208 sec. in contact with the wire in the plutonium experiment. The total amount swept out can be calculated from the time spent in contact with the wire and the known amount collected according to the following formula:

$$A = A_0 (1 - e^{-\lambda t})$$

where A = activity collected on wire.

A_0 = total activity swept out of solution.

t = time of contact with the wire (time of traversing the 10 ft. aluminum collecting tube).

$\lambda = 0.693/T_{1/2}$ of gaseous parent.

The estimated errors in Table III are only to indicate the spread of the different analyses from the average.

Analysis of the decay curves into the various components gave the known half-lives for the various isotopes within experimental error. Where

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possible the counts per minute were read from the graph at a point near the middle of the curve which would be relatively insensitive to the slope and therefore to the actual half-life obtained by subtraction of other components. The estimates of the precision indicate that the data are good.

To estimate the accuracy of the figures given for the per cent of a chain swept out it would be necessary to repeat the experiments in the same apparatus and under the same conditions in order to determine the reproducibility of the data.

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Table II.

Uranium Experiment

13 cc. solution containing 1.00 grams U.
 Bombardment 5/31/45 from 14:10 to 18:10 at 300 EV
 Flow rate = 338 cc/min. Length of wire = 10 ft.
 Inside diam. of aluminum collecting tube = 1 inch
 Spray = $10^{-4}\%$

| Sample | Cpm at time given | Chem. yield | Aliquot | Average Cpm at time giv. | % swept out | Remarks |
|-----------|-------------------|-------------|--------------------|--------------------------|-------------|--|
| S5 min Ba | 1st shelf | | | | | |
| S-1 | 2230 at 6/1 00:00 | 19.0/35.5 | 2×10^{-4} | | | OH ⁻ ppt. at 22:21 5/31 Cpm read from graph after subtracting 12.5d tail of Ba ¹⁴⁰ Factor $1-e^{-\lambda t}$ to correct W to total amount swept out = .0.962 |
| S-2 | 1300 " | 10.7/35.5 | " | 2.186×10^7 | | |
| S-3 | 2850 " | 21.9/35.5 | " | | | |
| S-4 | 2750 " | 20.8/35.5 | " | | | |
| W-1 | 686 " | 17.9/35.5 | 10^{-4} | 1.38×10^7 | 39.1% | |
| W-2 | 510 " | 12.9/35.5 | " | | | |
| W-3 | lost | - | " | | | |
| W-4 | 520 " | 22.3/35.5 | " | | | |
| 3.5 hr Y | 2nd shelf | | | | | |
| S-1 | 620 | 2/26.22 | 4×10^{-5} | | | Cpm read from graph after subtracting 11 hr. tail. 2.3d tail of 39 also subtracted first from S samples. |
| S-2 | 1160 | 2.8/26.22 | " | 2.63×10^8 | | |
| S-3 | 940 | 2.4/26.22 | " | | | |
| S-4 | lost | | " | | | |
| W-1 | 8300 | 7.4/26.22 | 4×10^{-3} | | 2.77 | |
| W-2 | 8500 | 7.5/26.22 | " | 7.51×10^9 | | |
| W-3 | bad curve | 6.6/26.22 | " | | | |
| W-4 | lost | | " | | | |

Table II.
Uranium Experiment

| Sample | Cpm at time given | Chem. Yield | Aliquot | Average Cpm at time giv. | % swept out | Remarks |
|----------|----------------------------|-------------|--------------------|--------------------------|-------------|--|
| 11 hr Y | Cpm 1st shelf 6/2 00:00 | | | | | |
| S-1 | 300 | 2/26.22 | 4×10^{-6} | } 1.35×10^8 | } 0.49% | |
| S-2 | 540 | 2.8/26.22 | " | | | |
| S-3 | 150 | 2.4/26.22 | " | | | |
| S-4 | lost sample | - | - | | | |
| W-1 | 780 | 7.4/26.22 | 4×10^{-3} | } 6.73×10^5 | | |
| W-2 | 750 | 7.3/26.22 | " | | | |
| W-3 | bad curve | 6.6/26.22 | " | | | |
| W-4 | lost | - | - | | | |
| 10 hr Sr | Cpm 6/1 18:00 1st shelf | | | | | |
| S-1 | 14,800 | 17.2/36.4 | 4×10^{-4} | } 6.28×10^7 | } 14.8% | |
| S-2 | 13,800 | 16.5/36.4 | " | | | |
| S-3 | 9900 | 6.2/36.4 | " | | | |
| S-4 | 6450 | 6.9/36.4 | " | | | |
| W-1 | 5200 | 13.3/36.4 | 10^{-3} | } 1.43×10^7 | | |
| W-2 | 2160 | 5.5/36.4 | " | | | |
| W-3 | lost | - | - | | | |
| W-4 | bad curve | 5.5/36.4 | " | | | |
| 17 hr Zr | Cpm 1st shelf 6/2 18:00 | | | | | |
| S-1 | 10,800 | 19.1/27.12 | 4×10^{-4} | } 3.83×10^7 | | 2.3d tail of 39 subtracted from 8 samples. |
| S-2 | 10,300 | 17.0/27.12 | " | | | |
| | | | | | 0.161% | |
| W-1 | 790 | 18.2/27.12 | 2×10^{-2} | } 3.79×10^6 | | |
| W-2 | 800 | 19.0/27.12 | " | | | |

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Table II.
Uranium Experiment

| Sample | Cpm at time given | Chem. Yield | Aliquot | Average Cpm at time giv. | % swept out | Remarks |
|-------------------------------------|----------------------------|-------------|--------------------|--------------------------|-------------|---|
| 33 hr Ce | Cpm 1st shelf 5/2 00:00 | | | | | 13.5 d Pr daughter activity subtracted from all S curves. 28 d tail subtracted from W curves. |
| S-1 | 10,300 | 9.7/24.6 | 2×10^{-4} | | | |
| S-2 | 4900 | 4.7/24.6 | " | 1.26×10^3 | | |
| S-3 | 9700 | 9.5/24.6 | " | | | |
| S-4 | 9500 | 9.8/24.6 | " | | | |
| W-1 | 440 | 9.5/24.6 | 2×10^{-2} | | 0.047% | |
| W-2 | 510 | 10.2/24.6 | " | 5.91×10^4 | | |
| W-3 | 430 | 9.2/24.6 | " | | | |
| W-4 | 450 | 9.2/24.6 | " | | | |
| 12.5 d Ba | Cpm 6/9 | | | | | |
| OH ⁻ ppt at 6/9 11:45 | | | | | | |
| S-1 | 8109 at 13:00 | 13.9/35.5 | 2×10^{-3} | 1.011×10^7 | | |
| S-2 | 4803 " 13:10 | 11.5/35.5 | " | | 23.1% | |
| W-1 | 2243 " 13:59 | 8.8/35.5 | 4×10^{-3} | 2.91×10^6 | | |
| W-2 | 3074 " 14:03 | 11.9/35.5 | " | | | |
| 55 d Sr | Cpm 1st shelf 6/16 | | | | | Factor $(1 - e^{-\lambda t})$ to correct W to total amounts swept out = 0.653 |
| S-1 | 5827 | 16.1/36.4 | 4×10^{-2} | 3.285×10^6 | | |
| S-2 | 5987 | 19.4/36.4 | " | | 61.5% | |
| W-1 | 1200 | 19.0/36.4 | 10^{-2} | 2.30×10^5 | | |
| W-2 | 1375 | 21.2/36.4 | " | | | |

Table II.
Uranium Experiment

| Sample | Cpm at time given | Chem. Yield | Aliquot | Average Cpm at time giv. | % swept out | Remarks |
|----------|--------------------|-------------|--------------------|--------------------------|-------------|--|
| 275 d Ce | Cpm 1st shelf 5/20 | | | | | Counts taken through 175 mg Al absorber. |
| S-1 | 2308 | 19.2/28.8 | 4×10^{-3} | 8.69×10^5 | | |
| S-2 | 2355 | 19.5/28.8 | " | | 0.052% | |
| W-1 | 63 | 19.9/28.8 | 2×10^{-1} | 4.48×10^2 | | |
| W-2 | 60 | 19.7/28.8 | " | | | |
| 28 d Ce | Cpm 1st shelf 7/1 | | | | | Cpm read from graph after subtracting 275 d Ce (17 min Pr) contribution. |
| | | | 4×10^{-3} | | | |
| S-1 | 7600 | 19.2/28.8 | " | 2.893×10^6 | | |
| S-2 | 7900 | 19.5/28.8 | " | | 1.75% | |
| W-1 | 7400 | 19.9/28.8 | 2×10^{-1} | 5.125×10^4 | | |
| W-2 | 6700 | 19.7/28.8 | " | | | |
| 60 hr Y | Cpm 1st shelf 7/8 | | | | | Chemical yield is overall yield for both Sr and Y. |
| S | 2138 | 61.3% | 0.25 | 1.395×10^4 | | |
| W | 1096 | 31.5% | 0.74 | 4.71×10^3 | 25% | |
| | Cpm 1st shelf 8/8 | | | | | |
| S | 1227 | 61.3% | 0.25 | 9.1×10^3 | | |
| W | 985 | 31.5% | 0.74 | 4.03×10^3 | 31% | |

Table II. Plutonium Experiment

13.0 cc. Solution containing 1.00 grams U as depleted oxide 3500:1
 Bombardment = 6/25/45 10:49 to 11:49 Inside diam. of aluminum collecting tube = 1 inch.
 Flow rate = 386 cc/min. at 500 KH. Spray = $2.5 \times 10^{-2}\%$
 Length of wire = 10 ft. Only 82.1% of Solution recovered.

| Sample | Cpm at time given | Chem. yield | Aliquot | Average cpm at time given | % swept out | Remarks |
|---|--------------------------|-------------|--------------------|---|-------------|---|
| S refers to Solution Samples. W refers to wire Samples. | | | | | | |
| 66 min Ba | Cpm let shelf | | | All S values have been divided by 0.821 | | 12.5d tail of Ba ¹⁴⁰ subtracted from curves. This value for % swept out takes into account the fact that 4% of the activity is due to U and is distributed according to results of U experiment; namely 47% is swept out and 53% is in solution. To get total swept out W must be divided by 0.97. This is factor $(1 - e^{-\lambda t})$ where $\lambda = \frac{0.693}{T_{1/2}}$; t = time spent by gas in ²³⁸ U gas 10 ft. Al. tube. $T_{1/2} = 41$ sec |
| S-1 | 5600 6/25 20:00 | 37.5/50.9 | 2×10^{-4} | 4.61×10^7 | 30.5% | |
| S-2 | lost | | | | | |
| W-1 | 13,200 " | 33.5/50.9 | 10^{-3} | 2.02×10^7 | | |
| W-2 | 10,750 " | 27.4/50.9 | | | | |
| 3.5 hr Y | Cpm let shelf | | | | | Values for counts per minute read from graph after subtracting 11 hour tail. A tail of 2.3d from 39 was also first subtracted from the S samples. |
| S-1 | 630 6/26/3:00 | 7.4/26.22 | 4×10^{-5} | | | |
| S-2 | 475 | 3.2/26.22 | | 1.308×10^8 | | |
| S-3 | 390 | 2.8/26.22 | | | | |
| S-4 | 340 | 4.7/26.22 | | | 1.17% | |
| W-1 | 2750 | 11.2/26.22 | 4×10^{-3} | | | |
| W-2 | 5100 | 12.6/26.22 | | 1.52×10^6 | | |
| W-3 | 2450 | 10.0/26.22 | | corrected for spray | | |
| W-4 | 2000 | 9.7/26.22 | | | | |
| 11 hr Y | Cpm let shelf 6/26/21:00 | | | | | |
| S-1 | 595 | 3.4/26.22 | 4×10^{-5} | | | |
| S-2 | 500 | 3.2/26.22 | | 8.15×10^7 | | |
| S-3 | 520 | 2.8/26.22 | | | | |
| S-4 | 410 | 4.7/26.22 | | | | |

Table II.
Plutonium Experiment

| Sample | Cpm at time given | Chem. yield | Aliquot | Average cpm at time given | % swept out | Remarks |
|----------|--------------------------------------|-------------|--------------------|---------------------------|-------------|---|
| 11 hr Y | Cpm 1st shelf 6/26/21:00 (Cont'd) | | | | | |
| | | | | | 0.27% | |
| W-1 | 420 | 11.2/26.22 | 4×10^{-5} | 2.10×10^5 | | |
| W-2 | 420 | 12.6/26.22 | | | | |
| W-3 | 235 | 10.0/26.22 | | corrected for spray | | |
| W-4 | 380 | 9.7/26.22 | | | | |
| 10 hr Sr | Cpm 1st shelf 6/26/18:00 | | | | | This value takes into account fact that 4% of the total activity comes from U and is distributed as in U experiment; namely 15% on wire and 85% in S. |
| S-1 | 12,700 | 17.2/36.4 | 10^{-3} | | | |
| S-2 | 14,600 | 19.8/36.4 | " | 3.45×10^7 | | |
| S-3 | 3700 | 4.3/36.4 | " | | | |
| S-4 | 4975 | 6.2/36.4 | " | | 8.9% | |
| W-1 | 35,500 | 21.2/36.4 | 2×10^{-2} | neglected, too low | | |
| W-2 | 29,500 | 19.8/36.4 | " | | | |
| W-3 | 18,500 | 8.6/36.4 | " | 3.45×10^5 | | |
| W-4 | 29,500 | 15.5/36.4 | " | | | |
| 17 hr Zr | Cpm 1st shelf 6/28/00:00 | | | | | |
| | | | 4×10^{-4} | | | |
| S-1 | 15,610 | 19.8/27.12 | " | 6.11×10^7 | | |
| S-2 | 15,300 | 19.3/27.12 | " | | | |
| | | | | | 0.032% | |
| W-1 | 430 | 19.1/27.12 | 2×10^{-3} | | | |
| W-2 | 520 | 18.4/27.12 | " | 1.55×10^4 | | |
| W-3 | 490 | 18.9/27.12 | " | corrected for spray | | |

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Table II.
Plutonium Experiment

| Sample | Cpm at time given | Chem. yield | Aliquot | Average cpm at time given | % swept out | Remarks |
|----------|-----------------------------|-------------|--------------------|---------------------------|-------------|--|
| 53 hr Ce | Cpm 1st shelf 6/27 00:00 | | | | | 13.5d Pr tail of daughter subtracted from S samples. 28d tail subtracted from W samples. |
| S-1 | 5900 | 8.9/24.6 | 2×10^{-4} | | | |
| S-2 | 6800 | 9.6/24.6 | " | 1.035×10^6 | | |
| S-3 | 8000 | 12.3/24.6 | " | | | |
| | | | | | 0.037% | |
| W-1 | 130 | 1.7/24.6 | 2×10^{-2} | | | |
| W-2 | 560 | 11.0/24.6 | " | 3.56×10^4 | | |
| W-4 | 515 | 10.6/24.6 | " | corrected for spray | | |
| 12.5d Ba | | | | | | GM ppt. at 15:01 6/27 |
| S-1 | 19,480 6/27/15:00 | 26.4/35.5 | 2×10^{-3} | 1.67×10^7 | | This result takes into account fact that 4% of total activity comes from U and is distributed 20% on wire and 80% in solution. Values not corrected for self-absorption or for L_{α} growth because of small change it makes in % swept out. Also not corrected for amount which escapes deposition since it amounts to only few %. |
| S-2 | 19,600 6/27/16:04 | 27.5/35.5 | " | | 11.0% | |
| W-1 | 4995 6/27/16:08 | 22.7/35.5 | 4×10^{-3} | 1.875×10^6 | | |
| W-2 | 4045 6/27/16:12 | 17.3/35.5 | " | | | |
| | | | | | | |
| 58d Sr | Cpm 2nd shelf 7/4 | | | | | This result takes into account the fact that 4% of total activity comes from U and is distributed 70% swept out, 30% in solution. Factor $(1-e^{-\lambda t})$ to correct W to total amount swept out = 0.606 |
| S-1 | 7397 | 24.5/36.4 | 10^{-1} | 1.315×10^6 | | |
| S-2 | 6236 | 21.4/36.4 | " | | 53.9% | |
| W-1 | 956 | 18.0/36.4 | 2×10^{-2} | 2.47×10^4 | | |
| W-2 | 1031 | 19.3/36.4 | " | | | |

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Table II.
Plutonium Experiment

| Sample | Cpm at time given | Chem. yield | Aliquot | Average Cpm. at time given | % swept out | Remarks |
|----------|---------------------------------------|-------------|--------------------|----------------------------|-------------|--|
| 284 Ce | Cpm 1st shelf 7/14 | | | | | |
| S-1 | 10,500 | 10.4/17.6 | 4×10^{-3} | 5.30×10^5 | | |
| S-2 | 11,100 | 11.5/17.6 | " | | | |
| | | | | | 0.7% | |
| W-1 | 4450 | 11.5/17.6 | 2×10^{-1} | 3.54×10^4 | | |
| W-2 | 4500 | 10.1/17.6 | " | Corrected for spray | | |
| 2764 Ce | Cpm 1st shelf 7/4 175 mg Al added. | | | | | Actually the spray should give more activity than was found on the wire for 2764 Ce. |
| S-1 | 1683 | 10.4/17.6 | 4×10^{-3} | 8.65×10^5 | | |
| S-2 | 1917 | 11.5/17.6 | " | | | |
| | | | | | 0% | |
| W-1 | 82 | 11.5/17.6 | 2×10^{-1} | corrected for spray | | |
| W-2 | 86 | 10.1/17.6 | " | = 0 | | |
| 57 hr Mo | Cpm 1st shelf 6/29 00:00 | | | | | |
| S-1 | 5750 | 17.5/40.6 | 4×10^{-4} | 4.52×10^7 | | |
| S-2 | 1850 | 4.6/40.6 | " | | | |
| | | | | | 0.042% | |
| W-1 | 190 | 12.2/40.6 | 2×10^{-2} | 1.91×10^4 | | |
| W-2 | 190 | 13.2/40.6 | " | corrected for spray | | |
| 60 hr Y | Cpm 1st shelf 7/8 | | | | | Chemical yield is overall yield for both Sr and Y. <u>SECRET</u> |
| S | 2100 | 65.2% | 0.25 | 1.29×10^4 | | |
| W | 940 | 49.2% | 0.53 | 3.3×10^3 | 20% | |
| | Cpm 1st shelf 5/15 | | | | | |
| S | 1100 | 63.2% | 0.25 | 8.33×10^3 | | |
| W | 790 | 49.2% | 0.53 | 2.84×10^3 | 24% | |

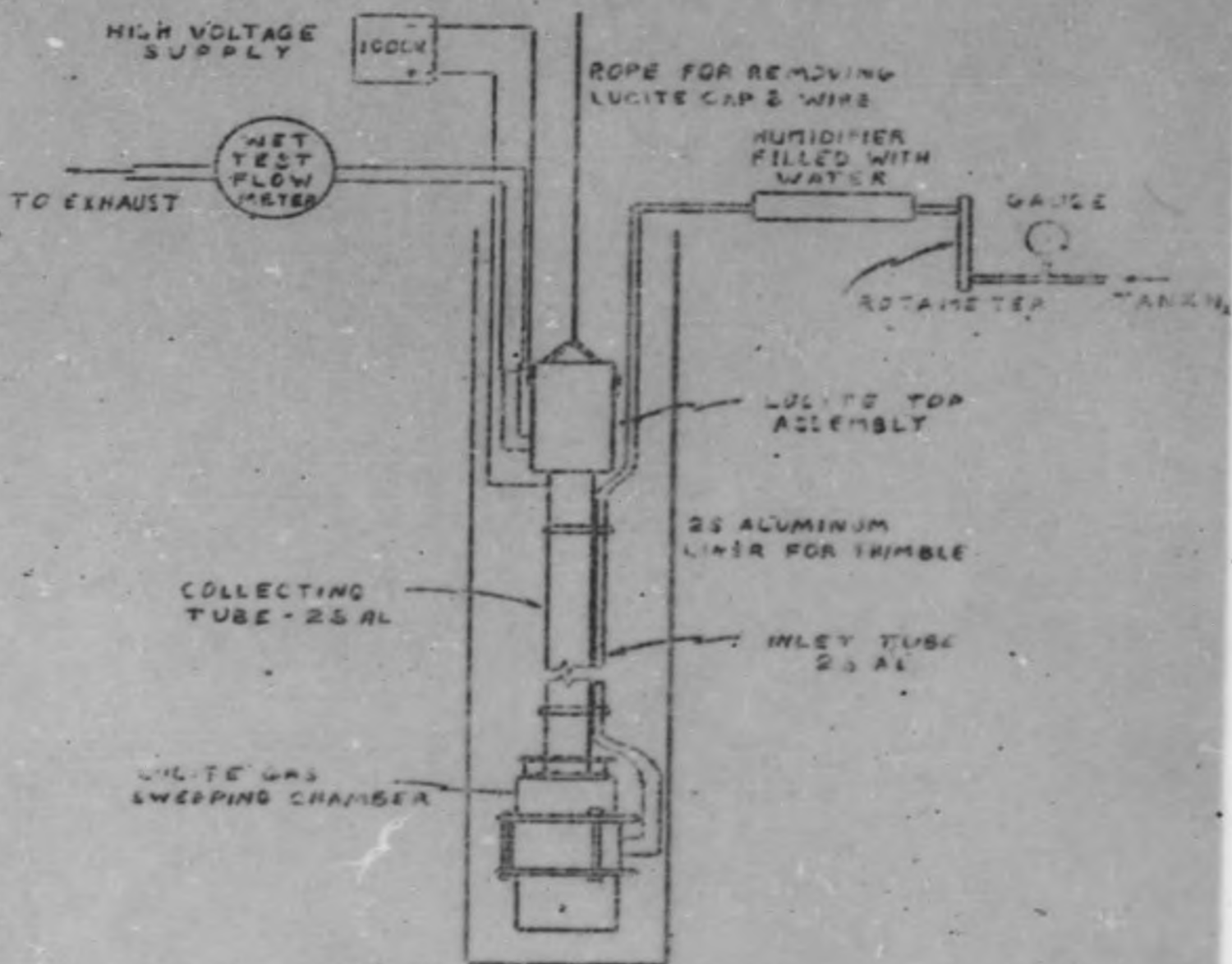
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Table III

% of Chain Swept out of Solution

| Chain Element Analyzed For | Uranium | Plutonium |
|----------------------------|----------------------------|---------------|
| 86 m Ba | 59.1 ± 1.5 | 50.5 ± 1.5 |
| 3.5 h Y | 2.77 ± 0.1 | 1.21 ± 0.25 |
| 11 h Y | 0.49 ± 0.04 | 0.27 ± 0.05 |
| 9.7 h Sr | 14.8 ± 0.7 | 8.9 ± 0.9 |
| 17 h Zr | 0.15 ± 0.002 | 0.034 ± 0.005 |
| 33 h Ce | 0.067 ± 0.004 | 0.035 ± 0.002 |
| 12.5 d Ba | 23.1 ± 0.8 | 11.0 ± 0.5 |
| 55 d Sr | 51.8 ± 0.7 | 53.9 ± 1.8 |
| 28 d Ce | 1.75 ± 0.1 | 0.7 ± 0.08 |
| 300 d Ce (17 m Pr) | 0.052 ± 0.002 | 0 |
| ~ 30 y Sr (60 h Y.) | 28 ± 3 | 22 ± 2 |
| 67 h Mo | 0 (from previous analyses) | 0.044 ± 0.011 |

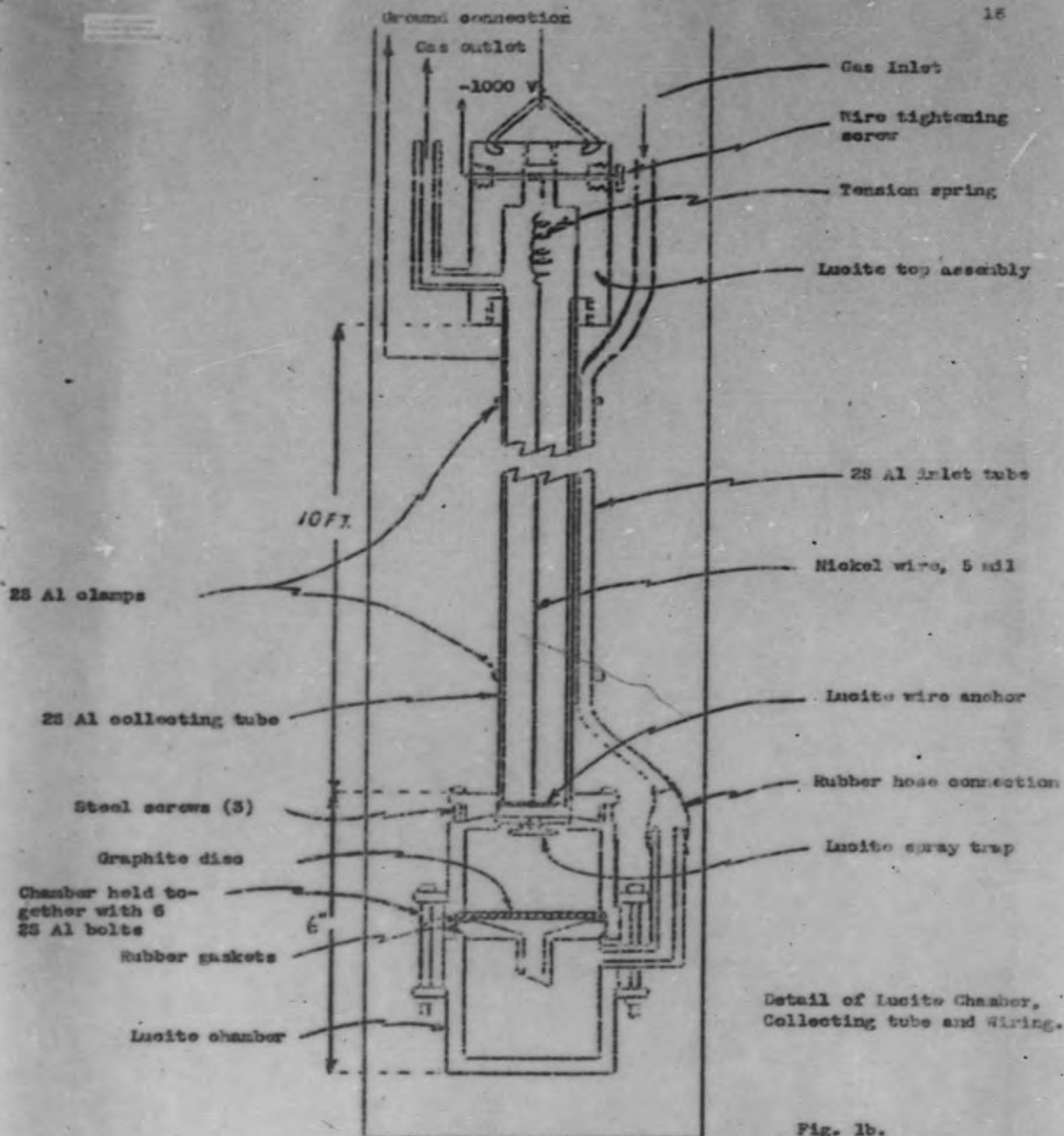
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Schematic Apparatus Assembly

Fig. 1a

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Addendum: Interpretation of and Inferences from these Experimental Results
William Rubinson

Table IV (which contains the data listed in Table III plus additional pertinent information) shows that the percentages of a gas swept from solutions of U and Pu increases with the average life of the gas, as is to be expected. But the average life of the gas cannot be the sole determining factor (assuming fixed flow-rate, bubble size, etc.), as is shown by the following facts:

1. The percentage of a given gas swept from U solution may be as much as five times that of the same gas swept from Pu solution. If average life of the gas were the sole determining factor, these percentages would be equal.

2. The percentages of 2.3s Kr⁽⁹²⁾ and 2.2s Kr⁹⁵ swept out of U solution differ by a factor of five, although the half-lives of these masses are almost exactly equal. The same is true of 1.4s Xe¹⁴¹ and 1.3s Xe¹⁴³. These pairs of gases show similar behavior in sweeping from Pu solution.

The explanation of these facts is to be sought in the variation of fission yield with atomic number. We have direct experimental evidence of the dependence of chain fission yield on chain member for several cases:

1. Hoagland and Sugarman (CC-3307) showed that in the chain
 $^{135}\text{Te} \xrightarrow{2m} ^{135}\text{I} \xrightarrow{8.7h} ^{135}\text{Xe} \xrightarrow{9.2h} ^{135}\text{Cs} \xrightarrow{2 \times 10^4 y} \text{stable Ba}^{135}$
 (Note: $^{135}\text{Xe} \xrightarrow{10m} ^{135}\text{I} \xrightarrow{\sim 10\%}$)
 about 5% of the total Xe¹³⁵ is produced independently of I¹³⁵

2. Goldstein, Schuman, and Rubinson (CN-2929) found that in the chain
 $^{141}\text{Xe} \xrightarrow{1.7s} \text{(short)} ^{141}\text{Cs} \xrightarrow{18m} ^{141}\text{Ba} \xrightarrow{3.5h} ^{141}\text{La} \xrightarrow{28d} \text{stable Pr}^{141}$
 about 20% of the Ce¹⁴¹ is produced independently of the Ba¹⁴¹.

3. L.K. Glendenin at Clinton has recently found (private communication) that in the chain

$^{83}\text{Se} \xrightarrow{25m} ^{83}\text{Br} \xrightarrow{2.4h} ^{83}\text{Kr} \xrightarrow{1.9h} \text{stable Kr}^{83}$

about half the Br⁸³ is produced independently of the Se⁸³.

There is also some indirect evidence for the dependence of fission yield on atomic number from Jentschke's measurements (Zeit.f. Physik, 120, 155 (1942)) of the energy associated with individual fission fragments. These show that a given pair of masses can be produced with different energies, and one explanation of this is that a given mass may be produced with different atomic numbers. Flammersfeld, Jensen, and Gentner (Zeit.f. Physik, 120, 450 (1942)) did experiments similar to Jentschke's. Their fig. 8 (p. 457) gives an idea of the wide variations in energy with which a given pair of masses can be produced in fission.

Now, suppose that for a given mass number, A, the independent fission yields of the different atomic numbers Z_A vary as represented schematically in Fig. 2, where a six-membered chain is assumed. The total yield of a given Z_A (i.e., the independent yield of that Z_A plus the independent yields of all the isotopes that precede it in the chain) as a function of Z_A is calculated simply from Fig. 2 and shown in Fig. 3. We make the reasonable assumption that in Pu fission, the curve of independent yield vs. Z_A is shifted one atomic number unit to the right with respect to the corresponding curve for U fission.

Since the "% swept out" listed in Table IV is really

$$\frac{\text{total gas yield} \times \text{fraction swept out}}{\text{total chain yield}} \times 100$$

and the "fraction", for a given set of experimental conditions, depends only on the average life of the gas, the differences in percent of a given gas swept out of U and Pu solutions is to be ascribed to the differences in the total yield of this gas in U and Pu. The manner in which this difference in total gas yields depends on the particular position that the gas member occupies in the chain can be seen from Fig. 3. If the gas occupies a position close to the stable member of the chain, the percentage swept out of U and Pu solutions can be expected to be approximately equal. According to Fig. 3, the ratio of total gas yield in U to that in Pu (both yields normalized to the same total chain yield) increases with separation of gas member from stable member. Actually, as is seen from Table IV, the situation is not that simple. In the schematic diagrams, no account is taken of the differences in the yield vs. Z_A curves for different mass numbers. For instance, for mass number 99 the Pu curve is wider than the U curve, while the opposite is true for mass number 144.

According to what has been said above, the ratio

$$R = \frac{\% \text{ swept out of U solution}}{\% \text{ swept out of Pu solution}}$$

can be written

$$R = \frac{\text{total gas yield in U}}{\text{total gas yield in Pu}} \times \frac{\text{total yield in Pu}}{\text{total yield in U}}$$

From the data in table IV and our knowledge of fission yields in U and Pu (the fission yields in Pu are provisional) we can determine the interesting ratio

$$\frac{\text{total gas yield in U}}{\text{total gas yield in Pu}}$$

The results of the calculation are given in Table V. Note the striking fact that although in most cases the fission yields in U and Pu differ by relatively small amounts, the total gas yield from U is in most cases more than twice as large as the total gas yield from Pu. This may be of some importance in the problem of the evaluation of radioactive gases from homogeneous piles.

Table IV

| Pass Number | Gas Member | Isotopes Analyzed | Number of β Decays from Gas to stable | β | β swept out* - Pu |
|-------------|----------------|----------------------|--|------------------|----------------------------|
| 89 | 2.6m Kr | 55d Sr | 3 | 51.8 \pm 0.7 | 57.5 \pm 1.8 |
| 90 | short Kr | \sim 36y Sr(80h Y) | 4 | 28 \pm 3 | 22 \pm 2 |
| (91) | 9.3s Kr | 9.7h Sr | 4 | 14.8 \pm 0.7 | 8.9 \pm 0.9 |
| (92) | 2.3s Kr | 3.3h Y | 4 | 2.77 \pm 0.1 | 1.21 \pm 0.25 |
| 95 | 2.2s Kr | 11h Y | 5 | 0.49 \pm 0.04 | 0.27 \pm 0.05 |
| 97 | short Kr | 17h Zr | 6 | 0.15 \pm .002 | 0.034 \pm .005 |
| 99 | short Kr | 67h Mo | 8 | 0 | 0.044 \pm 0.011 |
| 139 | 41s Xe | 86m Ba | 3 | 39.1 \pm 1.6 | 30.5 \pm 1.5 |
| 140 | 16s Xe | 12.5d Ba | 4 | 23.1 \pm 0.8 | 11.0 \pm 0.5 |
| 141 | \sim 1.4s Xe | 28d Ce | 5 | 1.75 \pm 0.1 | 0.7 \pm 0.08 |
| 143 | \sim 1.3s Xe | 33h Ce | 6 | 0.047 \pm .004 | 0.035 \pm .002 |
| 144 | short Xe | 275d Ce(17m Pr) | 6 | .052 \pm .002 | 0 |

These data are copied from Table III

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Table V

| Gas | K | Total chain yield (%) | | $\frac{\text{total gas yield in U}}{\text{total gas yield in Pu}}$ |
|---------------------------|------|-----------------------|-------------------|--|
| | | U ^a | Pu ^{a,b} | |
| 2.6m Kr ⁸⁹ | 0.97 | 4.6 | 1.83 | 2.42 |
| (short) Kr ⁹⁰ | 1.27 | (5.2) | (2.4) | 2.75 |
| 9.3s Kr ⁽⁹¹⁾ | 1.66 | 5.9 | 2.72 | 3.84 |
| 2.3s Kr ⁽⁹²⁾ | 2.28 | 5.1 | (4.6) | 2.52 |
| 2.2s Kr ⁽⁹⁵⁾ | 1.8 | 6 | 6.16 | 1.75 |
| (short) Kr ⁹⁷ | 4.42 | 6 | (6.3) | 4.2 |
| (short) Kr ⁹⁹ | 0 | 6.8 | 6.2 | 0 |
| 41s Xe ¹³⁹ | 1.28 | 6.3 | (6.1) | 1.32 |
| 16s Xe ¹⁴⁰ | 2.1 | 6.1 | 5.0 | 2.56 |
| ~ 1.4s Xe ¹⁴¹ | 2.5 | 5.7 | 4.45 | 3.2 |
| ~ 1.3s Xe ¹⁴³ | 1.24 | 5.4 | 4.68 | 1.43 |
| (short) Xe ¹⁴⁴ | ∞ | 5.0 | 3.45 | ∞ |

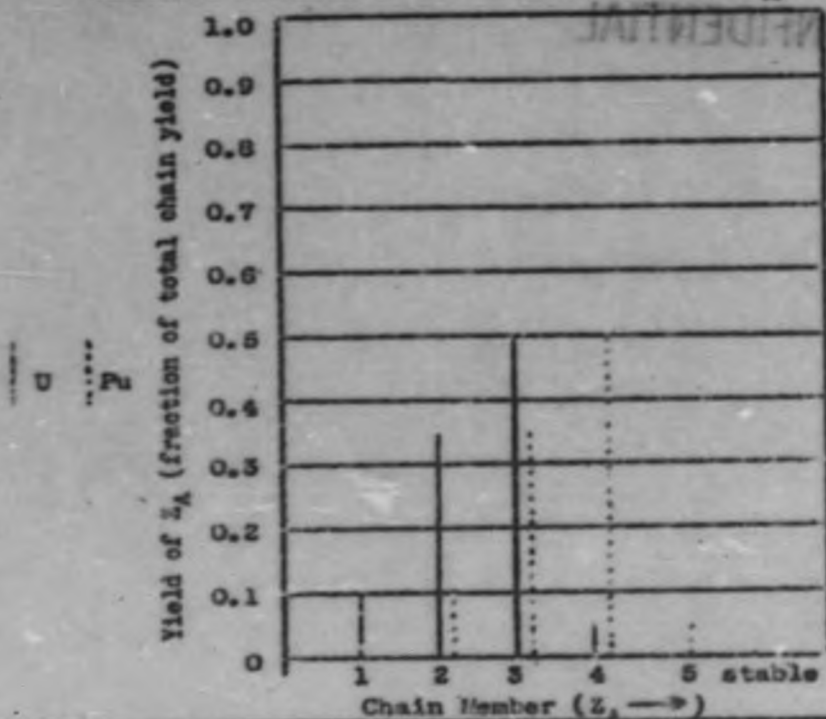
^a Yields taken from the latest compilation, CL-CDC #8

^b Yields taken mostly from CN-2799. The 140 yield is from recent unpublished work. The yields in parenthesis are estimates from a yield vs. mass curve. with the exception of the 99 and 140 chains, all these yields may be 15% high.

END

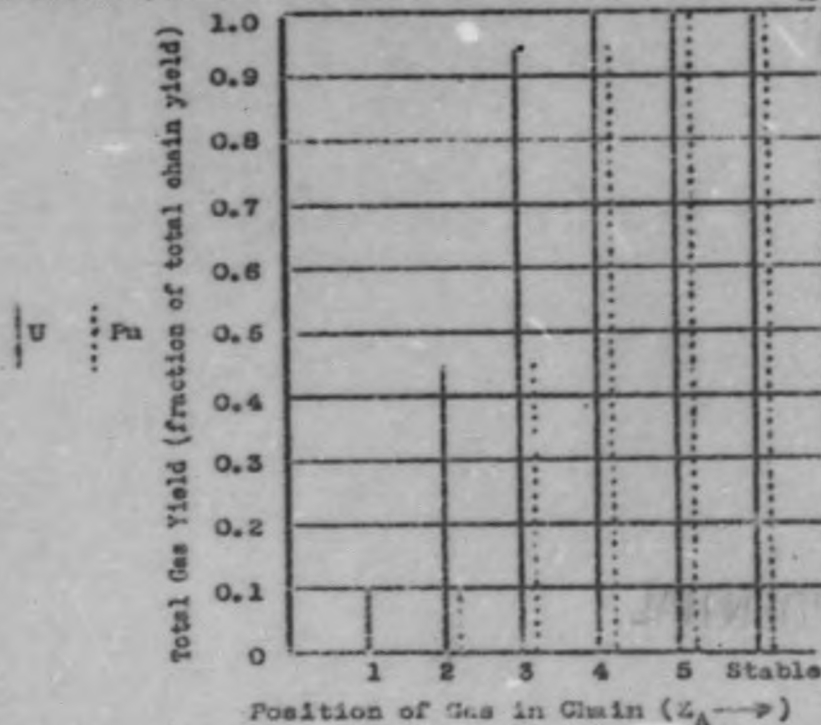
Fig. 2

Schematic Dependence of Fission Yield on Z_A for U and Pu



Schematic Dependence of Total Gas Fission Yield on Z_A of Gas Member

for U and Pu



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