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# EVALUATION OF A 4π NEUTRON COUNTER FOR THE NONDESTRUCTIVE ANALYSIS OF URANIUM-234 IN URANIUM HEXAFLUORIDE

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FEBRUARY 1976

UNION CARBIDE

OAK RIDGE GASEOUS DIFFUSION PLANT
OAK RIDGE, TENNESSEE

*prepared for the* **U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION**
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Oak Ridge Gaseous Diffusion Plant
Union Carbide Corporation
Oak Ridge, Tennessee

Prepared for the U.S. Energy Research and Development Administration under U.S. Government Contract W-7405 eng 26

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## ABSTRACT

A neutron counter has been evaluated for the nondestructive analysis of uranium-234 in uranium hexafluoride. The instrument was designed by Los Alamos Scientific Laboratory for the Nuclear Safeguards Program. The method assumes that the only source of neutrons is from $^{19}F(\alpha\ n)^{22}Na$ reaction from uranium-238, uranium-235, and uranium-234, the yield from uranium-234 being the most abundant.

This evaluation shows that the mean of the difference between the $4\pi$ neutron counting method and mass spectrometry is 0.0002 wt % uranium-234 ± 0.00015 at the 95% confidence level. However, the presence of uranium-232 produces significant errors in the system. If the neutron technique is to be used for samples containing uranium-232 in equilibrium with its daughters, the 2.61-MeV gamma peak of thallium-208 must be measured and used to correct for the uranium-232.

It has been postulated that the neutron counter might also be used to relate the uranium-234 content of a sample to the uranium-235, thus estimating the uranium-235 concentration. This evaluation notes, however, that in a variety of uranium feeds and enriched product, the uranium-234/uranium-235 ratio varies too widely to enable a valid uranium-235 determination.

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# EVALUATION OF A $4\pi$ NEUTRON COUNTER FOR THE NONDESTRUCTIVE ANALYSIS OF URANIUM-234 IN URANIUM HEXAFLUORIDE

## INTRODUCTION AND SUMMARY

The neutron counter was set up at the Oak Ridge Gaseous Diffusion Plant (ORGDP) for the purpose of evaluating it in an independent measurement of uranium-234 in uranium hexafluoride ($UF_6$) from an experiment in the Minor Isotopes Safeguards Technique (MIST).

Sixteen 2S cylinders from the MIST test containing $UF_6$ that had been routinely analyzed for uranium-234 and uranium-235 by mass spectrometry were measured in the neutron counter. One cylinder was measured 17 times to establish the precision for the instrument. Each cylinder was measured for 1000 sec, followed by a 1000-sec background count. The counts averaged between 25,000 and 45,000 counts for 1000 sec, and the background ranged from 450 to 750 counts for the same counting period. Each 2S cylinder contained approximately 1700 g of $UF_6$.

## INSTRUMENTATION

The detector consists of fourteen helium-3 tubes, embedded in a polyethylene cylinder (60 cm in diameter by 79 cm in height) approximately the size of a 55-gal drum. A well inside the detector assembly will accept 1S and 2S cylinders. A 1S cylinder is 4 cm in diameter, and has a fill limit of 454 g of $UF_6$, whereas a 2S cylinder is 9 cm in diameter and has a fill limit of 2225 g of $UF_6$ (see Figure 1).

## DISCUSSION AND RESULTS

The uranium-234 analysis is derived from a background-corrected neutron count, using the expression

$$\text{Wt \% Uranium-234} = aS - b,^*$$

where:

> S is the number of neutrons for 1000 sec/g $UF_6$ and the constants, a and b, are obtained from a best fit curve of plotted mass spectrometer data.

*Stieff, L. R., Walton, R. B., Reilly, T. D., Fields, L. W., Walker, R. L., Mullins, W. T., and Thoms, J. I., *Neutron Measurements of U-234 Isotopic Abundances in $UF_6$ Samples,* orally presented at the Institute of Nuclear Materials Management Meeting at New Orleans, Louisiana, June 1975.

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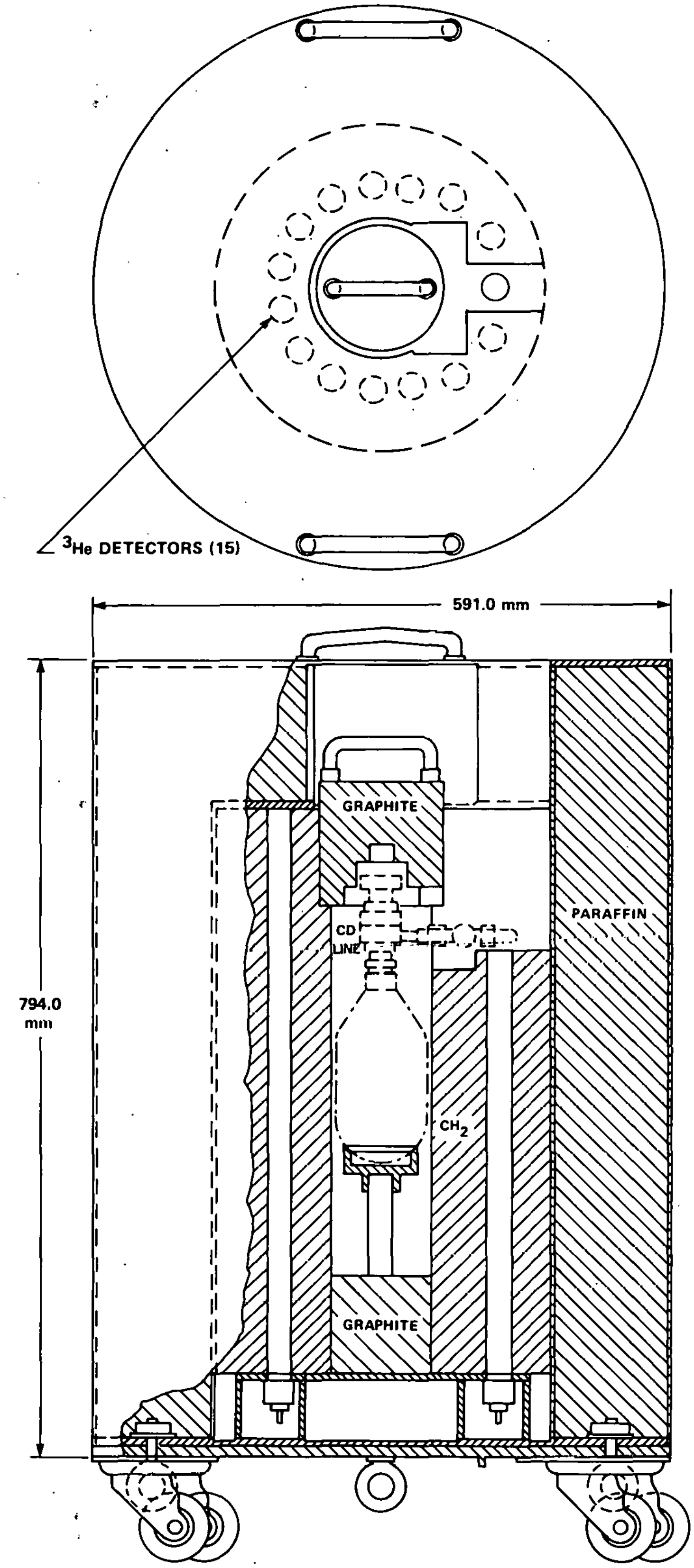


Figure 1
4$\pi$ NEUTRON COUNTER

This expression assumes that there are three sources of neutrons: uranium-234 (the most dominant in enriched uranium), uranium-235, and uranium-238 from the $^{19}F(\alpha\ n)^{22}Na$ reaction. Table 1 lists the neutron yields from the fluorides of the uranium isotopes.

Table 1[a]

($\alpha$ n) YIELDS FROM FLUORIDES

| Material | Yield, neutrons/sec/kg |
|---|---|
| $^{234}UF_6$ | $(5.76 \pm 0.42) \times 10^5$ |
| $^{235}UF_6$ | $122 \pm 9$ |
| $^{236}UF_6$ | $(3.95 \pm 0.29) \times 10^3$ |
| $^{238}UF_6$ | $27.9 \pm 2$ |

[a]Sampson, Thomas E., "Neutron Yields From Uranium Isotopes in Uranium Hexafluoride," *Nuclear Science and Engineering, Vol. 54*, p. 470 (August 1974).

It is estimated that the neutron yield per kg for uranium-232, without its daughters, is $3 \times 10^9$.* In equilibrium with its daughters, the neutron yield would be increased by a factor of 5.

Sixteen 2S cylinders containing $UF_6$ that had been routinely analyzed for uranium-234 and uranium-235 by mass spectrometry were obtained and measured in the neutron counter. Five cylinders were selected for use as standards. The $UF_6$ from three cylinders was reanalyzed for wt % uranium-234 at ORGDP, and $UF_6$ samples from five cylinders were analyzed at the Oak Ridge National Laboratory (ORNL). Table 2 shows the results. The averages of the wt % uranium-234 results from ORGDP and ORNL analyses were used to determine the a and b constants. The following equation was used to calculate the wt % uranium-234 by the neutron measurement:

$$\text{Wt \% Uranium-234} = 0.001347\ S - 0.004212$$

Table 3 shows the results.

*Stieff, L. R., et al., *loc. cit.*

Table 2

WEIGHT PERCENT URANIUM-234 BY MASS SPECTROMETRY AT ORGDP AND ORNL

| Cylinder No. | 210957 | 210967 | 964908 | 210913 | 210952 |
|---|---|---|---|---|---|
| ORGDP: | 0.02175 | 0.03194 | 0.01547 | | |
| | 0.02179 | 0.03151 | 0.01514 | | |
| | 0.02190 | 0.03148 | 0.01525 | | |
| | 0.02192 | 0.03136 | 0.01522 | | |
| | | | 0.01522 | | |
| Average | 0.02184 | 0.03157 | 0.01526 | | |
| ORNL: | 0.02156 | 0.03112 | 0.01552 | 0.01716 | 0.02748 |
| Best Value | 0.02170 | 0.03134 | 0.01539 | 0.01716 | 0.02748 |

The precision for a single measurement of the ORGDP results is ±0.00048, at the 95% confidence level. The ORNL results are reported to be ±0.0001.

Table 3

WEIGHT PERCENT URANIUM-234 BY MASS SPECTROMETRY AND 4π NEUTRON ANALYSIS

| Cylinder No. | S n/1000 sec per g $UF_6$ | Wt % Uranium-234 Neutron Analysis (N) | Wt % Uranium-234 Mass Spectrometry (MS) | MS-N |
|---|---|---|---|---|
| 210951 | 23.24 | 0.027 | 0.028 | 0.001 |
| 210959 | 18.86 | 0.021 | 0.022 | 0.001 |
| 490060 | 21.32 | 0.024 | 0.023 | -0.001 |
| 210968 | 26.07 | 0.031 | 0.031 | 0 |
| 964904 | 25.35 | 0.030 | 0.031 | 0.001 |
| 964905 | 24.94 | 0.029 | 0.029 | 0 |
| 210958 | 19.14 | 0.022 | 0.022 | 0 |
| 210961 | 19.38 | 0.022 | 0.023 | 0.001 |
| 210966 | 26.36 | 0.031 | 0.030 | -0.001 |
| | | | | $\bar{X}$ 0.0002 |

Note from Table 3, the mean of the difference between the two measurements is 0.0002 wt % uranium-234 ±0.00015 at the 95% confidence level.

## INTERFERENCES

The method assumes that since the neutron yield from uranium-234 is the most abundant, the method is relatively insensitive to changes in the uranium-234/uranium-235 ratio. The analyses of wt % uranium-234 and wt % uranium-235 in 438 $UF_6$ samples were reviewed, and Table 4 shows fluctuations typical of the uranium-234 to uranium-235 ratios.

Table 4

VARIATION OF URANIUM-234 IN URANIUM HEXAFLUORIDE

| Material | Uranium-234/Uranium-235 Ratio Range | | | High/ Low | No. of Samples |
|---|---|---|---|---|---|
| | Mean | High | Low | | |
| Reactor Processed Feed | 0.00814 | 0.01201 | 0.00467 | 2.572 | 96 |
| >3 Wt % Uranium-235 Product | 0.00873 | 0.00954 | 0.00736 | 1.296 | 193 |
| 2.4 to 2.8 Wt % Uranium-235 Product | 0.00836 | 0.0133 | 0.00672 | 1.979 | 149 |

Note from Table 4 the fluctuations of the uranium-234 to uranium-235 ratio in an extreme case can vary as much as 2.57 times. If this procedure is used to determine uranium-234 as a measure of the uranium-235, an error can be as large as 52% of the uranium-235 value.

Uranium-232, sometimes found in reactor-processed uranium, also interferes in this procedure. Two cylinders containing uranium-232 were analyzed by the neutron method. Table 5 shows the results.

Table 5

VARIATION OF URANIUM-232 IN REACTOR-PROCESSED URANIUM

| Material | ppb $\frac{\text{Uranium-232}}{\text{Uranium-235}}$ | Wt % Uranium-234, Neutron (N) | Wt % Uranium-234, Mass Spectrometry (MS) | $\frac{N - MS}{M}$, % |
|---|---|---|---|---|
| 490044 | 54 | 0.019 | 0.015 | 27 |
| 490045 | 29 | 0.013 | 0.011 | 18 |

Note that by the neutron method, only 29 ppb of uranium-232/uranium-235 will cause the wt % uranium-234 measurement to be erroneously high by 18%.

Using the neutron yields in Table 1, and assuming a $UF_6$ sample enriched to 1.5 wt % uranium-235, containing 0.015% uranium-234 and 54 ppb uranium-232/uranium-235, the calculations in Table 6 indicate that the uranium-232 produces 12% of the total number of neutrons.

Table 6

URANIUM-232 CALCULATED IN REACTOR-PROCESSED URANIUM

| Uranium Isotope | % Uranium Isotope | Neutron Yield | No. Neutrons | % of Total |
|---|---|---|---|---|
| Uranium-238 | 98.49 | 27.9 | 2748 | 20.83 |
| Uranium-235 | 1.50 | 122 | 183 | 1.39 |
| Uranium-234 | 0.015 | $5.76 \times 10^5$ | $8.64 \times 10^3$ | 65.50 |
| Uranium-232 | $5.4 \times 10^{-7}$ | $3 \times 10^9$ (a) | $1.62 \times 10^3$ | 12.28 |

(a) Estimation of neutron yield, not in equilibrium with daughters.

If the uranium-232 is in equilibrium with its daughter thallium-208, the 2.61 MeV of thallium-208 can be measured by gamma spectrometry, and used to correct the uranium-234 analysis.

## PRECISION

In order to check the precision of the instrument, one cylinder (490045) was measured 17 times over a period of several days. Table 7 shows the results.

Table 7

COUNTS/GRAM OF URANIUM HEXAFLUORIDE FOR 1000 SECONDS

| | | |
|---|---|---|
| 12.93 | 13.15 | 12.93 |
| 13.09 | 12.92 | 13.06 |
| 12.93 | 13.07 | 12.96 |
| 12.85 | 12.82 | 12.95 |
| 12.89 | 12.96 | 13.00 |
| 13.00 | 13.00 | - |

The mean of these measurements is 12.97 ± 0.045 at the 95% confidence level.

## CONCLUSIONS

It is concluded from this evaluation that a 4π neutron counter provides a fast, nondestructive method for the analysis of uranium-234 with good precision. From a study of nine 2S cylinders of $UF_6$, the mean of the difference between the 4π neutron method and mass spectrometry is 0.0002 wt % uranium-234 ±0.00015 at the 95% confidence level.

Two serious errors can affect the procedure, unless one applies some prior knowledge of the history of the sample, or makes supplementary corrective analyses. First, if the procedure is to be used to determine uranium-234 as a measure of the uranium-235, then as has been pointed out from Table 5, it is possible for the uranium-234/uranium-235 ratio to vary as much as 2.57 times and cause an error as large as 52%. From Table 6, it is also shown that the presence of uranium-232 as low as 29 ppb (on a uranium-235 basis) can cause an error as large as 18% in the uranium-234 analysis.

## ACKNOWLEDGMENTS

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