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MEASURING FISSION PRODUCT ACTIVITY IN
URANIUM CONTAINING U^{232} DAUGHTERS

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OAK RIDGE GASEOUS DIFFUSION PLANT

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ACTIVITY IN URANIUM CONTAINING
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Author: A. R. Flynn

A B S T R A C T

The method specified by the USAEC for measuring fission product activity in uranium hexafluoride has been adapted to material produced from spent nuclear fuel containing U^{232} and its daughters; a convenient gamma spectrometer correction for the daughter activity is used.

The uranium and thorium are extracted into tributyl phosphate, leaving the fission products and the U^{232} daughters (except Th^{228}) in the aqueous phase. The total gamma activity of the aqueous phase is measured in a high pressure ionization chamber as specified and corrected for the U^{232} daughter activity, using a correlation between the gamma activity of Tl^{208} (a U^{232} daughter) and the total gamma activity of the U^{232} daughters. To establish this correlation, the uranium is extracted from U^{232} standards, the remaining Tl^{208} is measured by gamma spectrometry and the total gamma activity is measured with the high pressure ionization chamber.

A similar correction is made for the beta activity of the U^{232} daughters, using a correlation established between the Tl^{208} gamma activity and the total beta activity of the U^{232} daughters.

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MEASURING FISSION PRODUCT ACTIVITY IN
URANIUM CONTAINING U^{232} DAUGHTERS

In the near future, uranium hexafluoride to be recovered from spent reactor fuels and fed into gaseous diffusion cascades will contain uranium-232 and its daughters which will interfere seriously with the measurement of the fission product activity in the feed. However, an effective, workable method of correcting for the U^{232} daughter interference has been devised to allow continued use of the relative, high pressure ionization chamber method specified by the United States Atomic Energy Commission for this measurement.

Using the specified analytical method, conformance to the specified activity levels is tested by extracting uranium and thorium from the fission products with tributyl phosphate, measuring the gamma activity of the fission products in a high pressure ionization chamber (HPIC) and correlating this measurement with a similar measurement of the gamma activity of aged natural uranium in equilibrium with Th^{234} (UX_1). In this separation of uranium and thorium from the fission products, any U^{232} daughters (except Th^{228}) remain in the aqueous phase with the fission products. The activity of the U^{232} daughters thereby introduces an error in the measurement of the fission product activity when the U^{232} content is significant. During the time the specified limit of U^{232}/U^{235} in the recovered fuel was only 11 ppb, the U^{232} posed neither specification nor analytical problems. The gamma activity from U^{232} and its daughters was insignificant. With changing designs and operating parameters for nuclear reactors, however, it has been necessary for the USAEC to raise the limit of U^{232}/U^{235} from 11 ppb to 110 ppb*. This report describes a method which has been developed to correct for errors caused by higher U^{232} concentrations. Continued use of the relative high pressure ionization chamber analytical method, with its advantages of convenience and direct relation of its readings to total radiation exposure, is thus permitted.

EQUIPMENT

The principal equipment used in making the measurements includes a high pressure ionization chamber to measure the total gamma activity from the fission products and U^{232} daughters, a gamma spectrometer to measure the activity in the 2.6 Mev gamma photo peak of thallium-208 and a beta proportional counter to measure the total beta activity from the fission products and U^{232} daughters.

* Federal Register, Vol. 32, No. 230, November 29, 1967.

PROCEDURE

In brief, the uranium hexafluoride is hydrolyzed to uranyl fluoride and the uranium and thorium are extracted by a method developed and used at the Oak Ridge Gaseous Diffusion Plant for several years. The uranium and thorium are extracted into a 50% tributyl phosphate-varsol solution, leaving the fission products and U^{232} daughters in the aqueous phase. The total gamma activity of the fission products and the U^{232} daughters is measured in the aqueous phase in the high pressure ionization chamber. In the recent adaptation of the method, the U^{232} daughter activity is then measured by gamma spectrometry and subtracted from the total gamma activity. The fission product beta activity is measured by counting aliquots of the aqueous solution in a proportional counter and correcting for the activity due to the U^{232} daughters.

Determination of the activity attributable to the U^{232} daughters, which is subtracted from the activity of the fission products plus U^{232} daughters, depends on using thallium-208 to represent the U^{232} daughters. The Tl^{208} activity is measured in the aqueous portion on a gamma spectrometer and correlated with a sample of known U^{232} daughter activity measured on the gamma spectrometer, the beta counter, and the high pressure ionization chamber. The procedural steps and calculations of the corrections can be seen in table I.

TABLE I

 U^{232} DAUGHTER ACTIVITY, CORRECTION FACTORS

<u>Known U^{232} daughter activity in standard, by HPIC, mv</u>	
Tl^{208} activity in standard, by γ spect. at 2.6 Mev, cpm	$= \gamma$ correction factor
<u>Known U^{232} daughter activity in standard, by β counting, cpm</u>	
Tl^{208} activity in standard, by γ spect. at 2.6 Mev, cpm	$= \beta$ correction factor
(Sample activity, by γ spect. at 2.6 Mev, cpm) (γ corr. factor) =	
equivalent HPIC reading in mv	
(Sample activity, by γ spect. at 2.6 Mev, cpm) (β corr. factor) =	
equivalent beta counter reading in cpm	

RESULTS

In a test of the effectiveness of the method, five uranyl nitrate solutions, each containing 5 grams of aged natural uranium, were processed according to the described procedure; the uranium and thorium extracted quantitatively into the organic phase as shown by the beta and gamma activities in the aqueous and organic phases in table II. All activity measurements are expressed as a percentage of the activity of natural uranium in equilibrium with Th^{234} . Precisions are stated at the 95% confidence level.

TABLE II

EXTRACTION OF URANIUM AND THORIUM (UX_1) INTO TRIBUTYL PHOSPHATE
(Percent of Activity of 5 g of Aged Natural Uranium)

Number	Gamma Activity		Beta Activity	
	Aqueous Phase	Organic Phase	Aqueous Phase	Organic Phase
1	0 ± 5	103 ± 10	1 ± 2	100 ± 10
2	2 ± 5	102 ± 10	0 ± 2	99 ± 10
3	-1 ± 5	100 ± 10	2 ± 2	101 ± 10
4	1 ± 5	98 ± 10	1 ± 2	98 ± 10
5	0 ± 5	97 ± 10	0 ± 2	102 ± 10

Aliquots were next taken from a nitric acid solution of U^{232} and each aliquot was processed according to the described procedure. The high pressure ionization chamber measurements shown in table III were made on the solutions before and after extraction, as a measure of the extraction efficiency.

TABLE III

GAMMA ACTIVITY FROM U^{232} DAUGHTERS BEFORE AND AFTER EXTRACTION
(Percent of Activity of 5 g of Aged Natural Uranium)

Aliquot No.	Original Solutions	Extracted Solutions	
		Aqueous Portion	Organic Portion
1	25 ± 5	24 ± 5	0 ± 5
2	25 ± 5	24 ± 5	2 ± 5
3	28 ± 5	26 ± 5	2 ± 5
4	27 ± 5	27 ± 5	-1 ± 5
5	27 ± 5	27 ± 5	0 ± 5

The U^{232} daughters, with the exception of Th^{228} , remained in the aqueous phase.

Calibration factors relating high pressure ionization chamber and gamma spectrometer measurements of known solutions were obtained by spiking four uranyl nitrate solutions, each containing 5 grams of uranium, with U^{232} having a known activity. Each solution was processed according to the procedures developed. The total gamma activity of each solution was measured in the high pressure ionization chamber, and the activity in the 2.6 Mev gamma photo peak was determined on the gamma spectrometer. The 2.6 Mev peak for Tl^{208} was chosen as an index to the total gamma activity of the U^{232} daughters. Activities equal to that added as U^{232} daughters were found in the aqueous phase as shown in table IV. Activity measurements are expressed as millivolts (mv) on the high pressure ionization chamber and as counts per minute (cpm) on the gamma spectrometer.

TABLE IV
GAMMA ACTIVITY FROM U^{232} DAUGHTERS MEASURED BY
HPIC AND GAMMA SPECTROMETRY

Solution Number	Original Activity, HPIC (mv)	Activity Extracted Into Aqueous Phase		γ Correction Factor mv/cpm
		HPIC (mv)	Gamma Spectrometer cpm, 2.6 Mev	
1	90	80	135	0.593
2	85	75	133	0.564
3	90	94	159	0.591
4	92	90	162	0.556
Mean				0.576 ± 0.025

Finally, four uranyl nitrate solutions were prepared and each was spiked with fission products (Ru^{106} and Cs^{137}) to a gamma activity of 225% of the activity of natural uranium in equilibrium with Th^{234} . U^{232} was also added to each solution, and the solutions were processed according to the procedure. Total gamma activity measurements on the aqueous phase were made on the high pressure ionization chamber and the gamma activity in the 2.6 Mev peak of thallium-208 was measured on the gamma spectrometer. The results are shown in table V. Activities are again expressed as mv on the high pressure ionization chamber and as counts per minute on the spectrometer.

TABLE V
FISSION PRODUCT ACTIVITY MEASUREMENTS,
CORRECTED FOR U^{232} DAUGHTER ACTIVITY

Solu- tion Number	(A) Total γ Activity, mv	(B) 2.6 Mev Photo Peak, cpm	(C) γ Activity, U^{232} Daughters (B x 0.576)	(D) γ Activity from F.P. (A-C)	F.P. γ Activity, %, Aged Natural U D x 100 Natural U Act.
1	699 $\pm$ 25	351 $\pm$ 40	202	497	221
2	702 $\pm$ 25	362 $\pm$ 40	208	494	220
3	707 $\pm$ 25	384 $\pm$ 40	221	486	216
4	715 $\pm$ 25	353 $\pm$ 40	203	512	228

CONCLUSION

The results obtained in this development work clearly show that gamma spectrometric methods can be used to provide necessary corrections to high pressure ionization data, thereby enabling the high pressure ionization chamber to be used to measure fission product activity, even in the presence of U^{232} and its daughters.

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