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UFCC Calibration Measurements for Bulk U_3O_8 Powder Samples

Howard Menlove and Carlos Rael

Abstract

The uranium UF_6 coincidence counter (UFCC) was designed to measure 5A/B cylinders containing UF_6 in the passive neutron self-interrogation mode. The purpose of the present report is to extend the UFCC capability to other sample types such as oxides and liquids that might be found during verification activities. A ^{252}Cf source was introduced into the detector to make the active mode measurements possible, because only the UF_6 samples have adequate neutron emission for self-interrogation. The sample enrichments varied from 0.72% to 91% to establish the shape of the calibration curve and the measurement uncertainties. The measurements included the singles, doubles, and triples rates. The time correlations between the ^{252}Cf source and the induced fission reactions reduced the statistical error in the doubles and triples rates to be small enough for useful measurements in 5-10 minutes.

Introduction

The UFCC was designed to measure the ^{235}U mass in UF_6 cylinders in the passive-neutron self-interrogation mode [1][2]. For the present experiments, the detector was modified to perform active interrogation by introducing a ^{252}Cf source that was placed directly under the sample. The purpose of the work is to extend the potential applications of the UFCC to ^{235}U samples such as oxides, metals, and liquids that are not composed of UF_6 , and therefore require an interrogating source to produce a measurable neutron signal. The measurements included the singles (S), doubles (D), and triples (T) rates, with calibration functions for each of these observables. Any one of the calibration curves could be used for measuring unknown samples and the combination can provide robustness to the measurements. This report describes the detector, the calibration standards, measurement procedures, calibration results, and the associated errors.

Time Correlated Interrogation Measurement Concept

The measurements for the fissile content in the uranium powder make use of the recently developed time correlated induced fission reaction method (TCIF) [3], where the sample is interrogated with neutrons from the ^{252}Cf source. However, the measured doubles and triples rates are enhanced because the induced fission reactions (IF) are time correlated with the ^{252}Cf spontaneous fission (SF) interrogation source. Thus, the trigger events that initiate the coincidence gates can originate from either the SF and/or the IF. This higher effective $\bar{\nu}$ is the basis of the improved precision in the TCIF method. The measured multiplicity rates (doubles and triples) increase with the effective neutrons per fission in the time gate. The $\bar{\nu}$ of the ^{252}Cf source is 3.76 and the induced fission $\bar{\nu}$ for ^{235}U is 2.44, so the combined effective $\bar{\nu}$ is higher than for random neutron interrogation; however, only a fraction of the time correlated neutrons are measured. This higher effective $\bar{\nu}$ significantly increases the multiplicity counting rates and reduces the statistical error.

There are three probabilities that come into play in the TCIF method:

1. The physical coupling between the ^{252}Cf and the sample for the IF
2. The probability of counting at least one of the IF neutrons in the detector
3. The probability of the ^{252}Cf background neutrons of being counted in the ^3He tubes

The first coupling probability is a function of the separation distance of the ^{252}Cf source to the sample, the poly moderator, the sample size, and potential absorbers of the thermal neutrons in the sample such as ^{238}U . This probability for a least one ^{252}Cf source neutron causing an IF we can define as P_{IF} . It is a variable that can be determined from physical standards or MCNP calculations.

The second probability for measuring at least one of the IF neutrons depends on the detector efficiency for neutrons born at the sample location. For the UFCC, the measured efficiency was 17.6% for the sample position. Thus, each of the 2.44 IF neutrons have a 17.6% chance of being detected, so the combined probability for the 2.44 neutrons that at least one of the neutrons is detected is equal to [3]

$$P_d = 1 - (1 - e)^{2.44} = 1 - (1 - 0.176)^{2.44} = 0.376$$

where e is the detection efficiency (0.176).

The third probability of measuring the ^{252}Cf background neutrons is a function of the neutron shielding for the ^{252}Cf source relative to the detectors. For the UFCC, the efficiency for the ^{252}Cf source in the bottom poly disc was 18.0% and $\bar{\nu}$ is 3.76. Thus, the probability of measuring at least one neutron from a single spontaneous fission event would be

$$P_{\text{cf}} = 1 - (1 - e)^{3.76} = 1 - (1 - 0.18)^{3.76} = 0.526.$$

The three probabilities all combine in determining the doubles rate boost from the TCIF method. If fast-neutron interrogation is used, the IF cross sections are small, so the probability for the IF is so small that the TCIF method does not provide a benefit. Thermal-neutron interrogation is used for the TCIF method to gain about two orders of magnitude in the ^{235}U reaction cross section (compared to fast neutrons), and to extend the TCIF method down to the $\sim 10 \text{ g } ^{235}\text{U}$ range.

Note that the ^{252}Cf spontaneous fission emits an average of 3.76 neutrons, and when more than one of these SF neutrons cause an IF, the correlated measured doubles and triples rates increase significantly regardless of the other two probabilities (items 2 and 3 above).

The TCIF method brings into play a longer die-away time than for the random neutron AmLi source interrogation. The combined time correlations of the SF and the IF include both the basic detector specific die-away time that is about 70 μs for the UFCC, but also the die-away time of the interrogation IF that is about 80 μs . Thus, to have our gate interval include both SF and correlated IF, we need to increase the gate to more than 100 μs . However, the statistical error is a function of the accidental counts (A) pileup, and a longer gate will make the A related error larger. An analysis of the data indicated that a gate length of 128 μs was near optimum.

There are neutron backgrounds that need to be subtracted from the active neutron measurement as follows:

1. Neutrons from the ^{252}Cf source (singles, doubles, and triples) with no ^{235}U
2. Neutrons from the room background (negligible for doubles and triples but not singles)
3. Passive neutrons from the uranium sample (negligible for oxides and metals)

For the room backgrounds, only the singles rate was significant where the room background singles rate was only 1.5% of the ^{252}Cf source rate that was used in the UFCC,

Detector description

The UFCC is a neutron well counter with one ring of twelve ^3He tubes within a polyethylene-moderator that contains a central sample cavity [2]. **Error! Reference source not found.** shows the detector with the side door closed (left) and with the door open (right). For the active mode measurements using a ^{252}Cf source, a HDPE disc insert was fabricated to hold the interrogation source in a horizontal position under the containers of U_3O_8 powder.

Fig. 1. The UFCC detector with the side door closed (left) and with the door open (right).

Sample Characteristics

The samples that were used to calibrate the UFCC were cans containing ~ 1000g of U_3O_8 powder with enrichments varying from natural at 0.72% to HEU at 91% ^{235}U . The sample parameters are listed in Table I.

Table I. Uranium oxide powder (UO_2) samples used in the UFCC calibration

Item ID.	Enrichment (%)	^{235}U Mass (g)
A1-1127-1	0.723	6.6
A1-1125-2	1.961	17.0
A1-1126-2	3.063	26.5
A-323-1	10.1	95.3
USIO-12	11.9	118
UIISO-13	13.1	130
UIISO-17	17.5	173
UIISO-27	27.1	264
UIISO-38	38	373
UIISO-52	52.5	516
UIISO-66	66.4	655
UIISO-91	91.4	903

Detector Parameters

To calibrate the UFCC for UO_2 bulk powder samples, we used the sample set shown in Table I. The cans of oxide powder were positioned in the bottom of the UFCC setting on top of the ^{252}Cf locator disc. The INCC detector parameters settings were as listed in Table II.

Table II. Detector parameters for the calibration measurements

Shift Register Type	JSR-15
Efficiency (with insert)	17.6%
Die-away time	75 μ s
High Voltage (V)	1680
DT Coeff. A	0.7
DT Coeff. B ($A^{2/4}$)	0.13
Multiplicity dead-time	500 ns
Gate	128 μ s
Pre-delay	2.0 μ s
Doubles gate fraction	0.6093
Triples gate fraction	0.3871

Room Backgrounds

The measurements were performed at Los Alamos, TA-66 in the School-House complex that has plutonium storage in the same room adding to the singles neutron background. Also, the high elevation (~ 7200 feet) results in cosmic-ray reaction source neutrons. The time-correlated double and triples background rates were found to be negligible compared with the IF rates from the ^{252}Cf .

The room backgrounds were measured on each of the measurement days and the results are given in Table III. We see that the average background rates were 41.6 cps, 0.059 cps, and 0.002 cps for the singles, doubles, and triples, respectively. Thus, the room backgrounds were stable and negligible for the measurements. Also, the extremely low level for the triples background made the triples rates for the samples useful for our low yield ^{252}Cf source. We note that a higher yield ^{252}Cf interrogation source would increase the neutron accidental rate that would be beneficial for the singles measurements but detrimental for the triples measurements. We note that the doubles rate percent error is almost independent of the interrogation source strength for singles rates above ~ 2000 cps because of the accidental rate increase.

Table III, UFCC room background measurements.

Date	count time [s]	Singles [cps]	Doubles [cps]	Triples [cps]
20.08.11	900	40.262	0.047	0.001
20.08.13	900	41.251	0.054	0.003
20.08.12	900	41.637	0.106	0.005
20.08.13	300	42.174	0.037	0.003
20.08.12	900	42.529	0.053	0.002
20.08.11	900	42.637	0.069	0.002

Sample Measurement Results

The measurements were made using multiple short cycles to determine the scatter from the repeat runs. Most of the calibration measurement were for 450-900s, and the reported error was from the data scatter in the gross rates.

Table IV lists the samples and the measured S, D, and T rates and the associated errors. The gross rates include the counts from the ^{252}Cf source. Whereas, the net rates have had the counts from the ^{252}Cf source subtracted from the gross rates, so that the zero ^{235}U mass counting rates are zero. Figure 2-4 show plots of the gross counting rates for the singles, doubles, and triples rates, where a logarithmic trend-line has been fit to the data. We see that the majority of the counts come from the spontaneous fission reaction neutrons in the ^{252}Cf interrogation source for the S and D. However, for the T rates, the time-correlated induced fissions are similar to the ^{252}Cf background. The statistical uncertainty related to the background rate is included in the measured scatter for the gross counts.

The net counting rates carry the statistical error related to the gross counting rates, so the percent error for the net rates is higher than for the gross rates. We note that the error in the ^{252}Cf background rate is small because of longer counting times for the source with no sample present.

Table IV. UFCC active neutron counting rates with ^{252}Cf source A7-866 (16750 n/s on 20.08.13)

Sample ID	^{235}U Mass [g]	Singles [cps]	Singles Error [cps]	Doubles [cps]	Doubles Error [cps]	Triples [cps]	Triples Error [cps]	net S [cps]	net D [cps]	net T [cps]
252Cf	0	2957	0.50	516	0.50	55	0.50	0	0	0
A1-1127-1	6.6	2977	2.64	529.2	1.63	61.07	1.91	20	13.2	6.1
A1-1125-2	17	3051	2.62	561.7	2.74	67.51	2.25	93.8	45.7	12.5
A1-1126-2	26.5	3091	3.18	588.1	2.57	73.16	2.09	133.9	72.1	18.2
A1-323-1	95.3	3251	3.20	680.9	3.11	100.75	2.52	294	164.9	45.8
UIISO-12	118	3280	3.11	686.3	3.32	95.94	2.48	323.2	170.3	40.9
UIISO-13	130	3286	3.01	694.2	1.89	100.73	1.38	329.2	178.2	45.7
UIISO-17	173	3321	4.56	713.6	1.85	104.89	2.12	364.4	197.6	49.9
UIISO-27	264	3385	3.75	753.4	3.70	115.33	2.87	428.1	237.4	60.3
UIISO-38	373	3408	3.99	780.1	2.78	126.12	2.74	450.9	270	71.1
UIISO-52	516	3457	5.11	840.4	3.77	135	3.13	499.9	310	80
UIISO-66	655	3464	2.97	845.9	2.85	145	2.84	530	329.9	90
UIISO-91	903	3525	3.01	873	2.94	158.74	3.13	568.4	357	103.7

The statistical errors in the gross rates are listed in Table IV for the singles, doubles, and triples rates for a 450 s measurement interval. We note that this statistical error would be reduced for longer counting times by the square root of the time compared with 450s. The absolute counting error for the gross rates is almost constant for the samples set because it is dominated by the ^{252}Cf source that was a constant for all of the samples. We note that the percent error tends to grow rapidly as the ^{235}U mass becomes small, and a practical lower limit for the ^{235}U mass would be ~ 10 g.

Figures 2-4 show the measured gross singles, doubles, and triples rates as a function of the sample ^{235}U mass. The gross rate data has been fit with logarithmic trend-lines as an eye-guide. The thermal-neutron interrogation shows a rapid increase in the response for the lower enrichment levels followed by a more gradual increase for the higher enrichments because of neutron self-shielding. For more typical samples that might be found in the field, Monte Carlo MCNP calculations would be used to establish the sample specific calibration curves.

Fig. 2 The gross singles rates for the samples listed in table IV.

Fig. 3 The gross doubles rates for the samples listed in table IV

Fig. 4. The gross triples rates for the samples listed in table.

Figures 5-7 show the net rates after the subtraction of ^{252}Cf source and neutron backgrounds. The trend-line fits using the logarithmic function are only for the eye-guide and do not fit well for extrapolated data regions. Thus, the calibration data needs to specify the range of validity. (More details on the mass range is given in the following section).

Fig. 5. The net singles rates for the samples listed in table IV where the ^{252}Cf rate has been subtracted.

Fig. 6. The net doubles rates for the samples listed in table IV where the ^{252}Cf rate has been subtracted.

Fig. 7. The net triples rates for the samples listed in table IV where the ^{252}Cf ratye has been subtracted.

We see that the net rates tend to flatten out above $\sim 200 \text{ g } ^{235}\text{U}$ because of thermal-neutron self-shielding in the samples. However, for the low mass samples ($< 100\text{g}$), the measured response increases rapidly with ^{235}U mass.

The calibration standards all had the same size and only the enrichment changed. The constant size and surface area led to calibration curves that are smooth and exhibit the self-shielding effect for the higher ^{235}U mass loadings. If the samples had a volume and surface area that increased with the ^{235}U mass, the calibration curves would not show the saturation above 100-200 g because of the increase in the fill height that would result in less neutron self-shielding per unit of mass. Calibrations with variable sample size and materials should be investigated in the future using MCNP modeling.

Limited Range Calibration Function

The present calibration measurements included enrichments ranging from natural uranium up to 91% ^{235}U . A more typical sample range would be all HEU with enrichments above 20% ^{235}U . The more limited enrichment range allows calibration curves with a better fit to the data. Figure 8 shows the data trend-line fit for samples in the mass range from 264-903 g ^{235}U . We see that the data fits the curve much better for the limited data range. All the measurements were for only 450s, and the small scatter about the calibration curves illustrates the good precision for the method. In all cases, the calibration range of validity should be specified with the calibration parameters.

Fig. 8. The net singles, double, and triples rates for samples in the limited range from 264-903 g ^{235}U to represent HEU samples.

We note that for in-field measurements, the samples would have different containers and sizes than were used for the present calibrations. Thus, the in-field measurements of a group of samples would provide a relative calibration for the group, and an absolute calibration could be performed at a later time based on MCNP modeling of the actual sample geometries. This same approach would be used for liquid and metal uranium samples.

Measurement Errors

The measurement error is a function of the ^{235}U mass and measurement time. Table V lists the sample masses and the measured statistical error in the singles, doubles, and triples rates for a 450s measurement interval. We note that for the higher mass values, these errors will increase significantly when the data is projected to the ^{235}U mass values because of the shape of the calibration curves. The calibrations are unique for all three variables (S, D, and T), so the measurement duplicity would provide a confirmation of the measured mass and the robustness of the result.

Table V. Counting rate percent errors for singles, doubles, and triples for 450s measurements using ^{252}Cf source A7-866 (16750 n/s on 20.08.13)

235U	Net S	Net S	Net D	Net D	Net T	Net T
Mass [g]	[cps]	error [%]	[cps]	error [%]	[cps]	error [%]
6.6	20	12.05	13.2	16.049	6.1	55.22
17	93.8	10.20	45.7	6.748	12.5	26.81
26.5	133.9	5.14	72.1	3.341	18.2	10.63
95.3	294	1.80	164.9	2.014	45.8	6.32
118	323.2	1.66	170.3	2.008	40.9	6.81
130	329.2	1.54	178.2	1.330	45.7	3.84
173	364.4	1.75	197.6	1.204	49.9	4.60
264	428.1	1.32	237.4	1.614	60.3	5.30
373	450.9	1.26	270	1.174	71.1	4.02
516	499.9	1.32	310	1.213	80	3.34
655	530	0.94	329.9	0.971	90	3.01
903	568.4	0.85	357	0.929	103.7	3.41

Limited Range Calibration Function and 235U Mass Error Analysis

For potential field use, the enrichment range of the measurements would likely be between 20-93 % ^{235}U , thus, we have focused on the corresponding calibration curves to this range. The singles and doubles curves shown in Fig. 8 would be the best choices because of they have smaller statistical errors than the triples curve. Table VI presents the singles and doubles errors for samples in this higher enrichment range. The reduced slope of the calibration curves is caused by thermal-neutron self-shielding that increases with enrichment, and the low slope of the calibration curves causes the error in the ^{235}U mass to be significantly greater than the errors in the net singles, and doubles rates as shown in Table VI.

The percent error in the 235U mass is smaller for the singles rate compared with the doubles rate, so that would suggest using the singles rates as the primary calibration and using the doubles rate in a confirmatory role.

We note that the neutron multiplication for the powder samples is very small compared with a similar mass in water such as the AEFC [4]. We also note that the present experiments used a very weak ^{252}Cf source (1.67e4 n/s), and ^{252}Cf sources that are an order of magnitude stronger at $> 1.0\text{e}5$ n/s are routinely used by the IAEA for detector calibration and in-field testing. For the singles measurements, a factor of 10 increase in the ^{252}Cf source strength would reduce the required measurement time by a factor of 10!

This same advantage is not present for the doubles statistical errors were the percent error does not change with increasing the interrogation source strength because of the accidental rate increase.

Table VI. The measured singles and doubles errors in the net rates and the calculated ^{235}U mass errors for a 1000s measurements using a ^{252}Cf source with a yield of 16750 n/s.

^{235}U Mass [g]	Net S error [%]	^{235}U error [%]	Net D error [%]	^{235}U error [%]
264	0.886	1.97	1.083	2.88
373	0.846	2.66	0.788	2.96
516	0.886	3.86	0.814	4.24
655	0.631	3.49	0.652	4.31
903	0.570	4.35	0.623	5.68

Summary

The UFCC was designed to measure 5A/B cylinders of UF_6 in the passive self-interrogation mode. The purpose of the present measurements is to extend the UFCC capability to other uranium sample types such as oxides, metals, and liquids that might be found during verification activities. A ^{252}Cf source was introduced to the detector to make the active mode measurements possible, because the uranium samples do not have enough neutron emission rate for a passive measurement.

The relatively low uncertainty of $\sim 3\text{-}20\%$ in the triples rates was a new finding in these measurements that was the result of using a rather low intensity ^{252}Cf interrogation source (16,750 n/s) so that the accidentals rate did not dominate the triples error. We note that the doubles and triples rates benefited significantly from the time correlated induced fission (TCIF) increase [3] in the correlated counting rates. The high nu-bar (3.76 n/fission) for ^{252}Cf source provided a significant probability of collecting two induced fission events inside the same time gate (128 μs). Potential future measurements using both a ^{252}Cf source and an AmLi neutron source for the interrogation could quantify the signal increase from the TCIF correlations.

The analysis of the three observables (S, D, and T) led to the conclusion (see Table VI) that the singles rate calibration provided the smallest statistical error in the ^{235}U mass value. Also, the singles rate can be easily increased by an order of magnitude by using a stronger ^{252}Cf source ($\sim 10^5$ n/s) that are typical of IAEA in-field neutron test sources. This would have the practical benefit of reducing the measurement error to $\sim 1\text{-}2\%$ in 5 min measurements. Future calibration data using a stronger ^{252}Cf source as well as MCNP simulations would be useful.

The calibration curves presented in this report correspond to thermal-neutron interrogation, so neutron self-shielding effects are strong, and the calibration results are dependent on the shape and surface area. For the measurement of different sample sizes and materials (powders, liquids, etc.), customized calibration curves would be required. These additional calibration curves can potentially be obtained via MCNP modeling before and/or after the field measurements. At the time of potential field measurements, the UFCC would provide a relative calibration for a group of samples. Later, the absolute calibration could be obtained based on the verification of any one of the samples by using an alternative measurement such as DA or by MCNP modeling.

The present measurements demonstrated that the UFCC has the capability to measure the ^{235}U mass in uranium samples similar to the active well coincidence counter (AWCC) [5] that is in routine use by the IAEA for the verification of bulk samples of ^{235}U . The UFCC has less efficiency than the AWCC, however, the UFCC is lighter and more portable for field applications. Also, the AWCC normally uses an AmLi neutron source for the active neutron mode interrogation. The present UFCC calibration made use of a ^{252}Cf source that is typical of sources that are used for field inspections to test the IAEA field equipment.

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