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**The Optimization and Calibration of the AWCC Using ^{252}Cf Interrogation and the
Comparison with an AmLi Neutron Source**

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1. BACKGROUND

The purpose of this report is to provide information on the modifications to the Active Well Coincidence Counter (AWCC) for the use of ^{252}Cf integration sources in place of AmLi sources [1]. Three AWCCs have been fabricated by commercial vendors for the future use at the China Safeguards Center of Excellence (COE). The AmLi neutron sources that have been used for the AWCC and other active neutron interrogation instruments are no longer available, and alternative neutron sources are needed. The ^{252}Cf sources provide the benefit that the interrogation source emits multiple neutrons per spontaneous fission that are time-correlated with the induced fissions (IF) in the sample. This increases the doubles signal and reduces the statistical error. The time correlated induced fission (TCIF) method [2] makes use of the ^{252}Cf sources that are readily available with the advantage of less initial cost, and a radioactive level in the tens of micro-curie range compared with the ^{241}Am neutron source in the curie range. However, the relatively short half-life of the ^{252}Cf source (2.64 years) will require a replacement after several years and that will cancel some of the cost advantage. The prior AWCC systems used two AmLi sources of ~ 1.1 Ci each to provide a neutron yield of $\sim 4.5 \times 10^4$ n/s each; whereas, a $10 \mu\text{Ci}$ ^{252}Cf source yields 4.4×10^4 n/s. For the present application, the ^{252}Cf source strength was $\sim 430 \mu\text{Ci}$ (on 10/1/2015), so the source will have an adequate neutron yield for AWCC applications for more than 15 years.

This report provides the AWCC detector modifications for using ^{252}Cf including detector parameters such as dead-time and die-away time, the efficiency, and the calibration coefficients. Also, included in the report is a direct measured comparison with the standard AmLi source (MRC-95) for the statistical performance.

2. TIME CORRELATED INTERROGATION CONCEPT

For the TCIF method, the sample is interrogated with neutrons from the ^{252}Cf source; however, the measured doubles rates are enhanced because the 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 ν is the basis of the improved precision in the TCIF method. The measured multiplicity rates (doubles and triples) increase rapidly with the effective ν (neutrons per fission in the time gate). The ν of the ^{252}Cf source is 3.76 and the induced fission ν for ^{235}U is 2.44, so the combined effective ν is

higher than for random neutron interrogation; however, only a fraction of the time correlated neutrons are measured. This higher effective ν 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 fissile mass, 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 modified AWCC, the measured efficiency was 38.2% for the sample position. Thus, each of the 2.44 IF neutrons have a 38.2% 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]

$$P_d = 1 - (1 - e)^{2.44} = 1 - (1 - 0.382)^{2.44} = 0.691,$$

where e is the detection efficiency.

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 modified AWCC, the efficiency for the ^{252}Cf source in the bottom poly end plug was 26.3% and ν is 3.76. Thus, the probability of measuring at least one neutron from a single spontaneous fission event would be

$$P_{\text{cf}} = 1 - (1 - e)^{3.76} = 1 - (1 - 0.263)^{3.76} = 0.683.$$

The three probabilities all combine in determining the doubles rate boost from the TCIF method. For the AWCC, the weak link in the combined probabilities is the first coupling probability because the relatively small sample only reacts with a minor fraction of the ^{252}Cf spontaneous neutrons because of its small size. 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 work except for multi-kg fissile samples. Thermal-neutron interrogation is used for the TCIF method to gain about 2 orders of magnitude in the ^{235}U reaction cross section (compared to fast neutrons), and to extend the TCIF method down to the $\sim 5 \text{ g } ^{235}\text{U}$ 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 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 AWCC, 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 (doubles and singles) with no ^{235}U
2. Neutrons from the room background (negligible for doubles but not singles)
3. Passive neutrons from the ^{235}U sample (negligible)

Only the first of these backgrounds is significant for the high yield ^{252}Cf sources that are used in the COE AWCCs.

The ^{252}Cf interrogation source background is impacted by the sample inside the detector because of neutron scattering from the sample and fast neutron fission in the ^{238}U . Thus, a dummy sample that contains no fissile mass was used for the background measurement. The ^{252}Cf background was measured overnight so that its statistical error was negligible. For the COE sample range (1-190 g ^{235}U) the ^{252}Cf source background is larger than the IF signal so care must be taken in measuring this background.

4. SYSTEM DESCRIPTION AND OPTIMIZATION

The AWCC shown in Fig.1 consists of a well type high density polyethylene moderator (HDPE) containing 42 ^3He tubes at 4 atm. pressure.[2] Measurements were made with both the ANTECH and Canberra AWCC systems with no significant differences in their performance. However, the ANTECH AWCC has an attachment to the electronics junction box that contained components to reduce the dead-time.

Fig. 1 Photo of one of the COE AWCC systems (left) with the ^3He tubes removed (right) to show the HDPE moderator design.

To obtain optimum performance of the AWCC, thermal-neutron interrogation was used. The modifications to the AWCC included, removing all Cd and the Ni annulus. The HDPE rings on both the top and bottom plugs were also removed to obtain a higher counting efficiency at the sample position. Figure 2 shows the ANTECH AWCC sample cavity with the Cd and HDPE rings removed from the bottom and top lid.

Fig. 2 Photo of the ANTECH AWCC system with a sample can in the cavity (left) and the ^{252}Cf source holder inserted in the end plug (right). The gray tape tabs were used to end plug and/or the ^{252}Cf source holder.

Fig. 3 Photo of the ^{252}Cf source holder showing the correct orientation of the components inside the holder where the screw-on lid is placed away from the sample.

Figure 3 shows the HDPE source holder for the ^{252}Cf where the orientation of the source and the small poly filler are important in positioning the source at the upper end of the holder. The position of the holder in the end plug has the HDPE screw threads away from the assay sample.

Measurements were made to determine the optimum position for the ^{252}Cf source inside the holder. The error in the doubles rate is a function of the neutron coupling to the sample as well as the neutron background in the ^3He detectors. To determine the optimum position for the source, the statistical error was measured at different depth positions for the ^{252}Cf inside the holder. The error was determined by the “sample mode” error evaluation in the INCC data collection software [4]. Each of the source configurations were measured for at least 30x20 s cycles and the spread in the doubles data was compared for the different positions. Figure 4 shows the doubles rate and error versus the ^{252}Cf source position in the end plug (cm), and we see that the error reduces as the source gets closer to the sample. We operated the system with the source at the minimum separations distance with the ^{252}Cf source at the top end of the holder.

Fig. 4 The net doubles rate and statistical error as a function of the ^{252}Cf source depth into HDPE end plug for a 600s measurement using a can of U_3O_8 powder.

The optimization measurements described above were used to determine that the mechanical condition of the AWCC that were as follows:

1. No Cd liners or Ni annulus
2. Both HDPE rings on the end plugs removed
3. The maximum sample cavity height was the standard 20cm
4. Only one ^{252}Cf source positioned in the bottom plug (i.e. no source in the lid)
5. The ^{252}Cf source holder was set flush with the bottom end plug with the orientation shown in Fig. 3.

To evaluate the change in the doubles rate and error as a function of the height of the can above the cavity bottom, measurements were performed for different can heights. Figure 5 shows the change in the doubles rate as a function of the sample height as well as the error in 600s for the different positions. The can used for the position optimization contained 186g of ^{235}U and had a diameter of 7.6cm and a height of 8.6cm containing a powder mass of 230g.

Fig. 5 The doubles rate and statistical error as a function of the height above the bottom for a 600s measurement using a can of U_3O_8 powder.

We see that the net doubles rate falls off slowly ($\sim 3\%/cm$) as the can is lifted above the bottom. The fast-neutron AWCC doubles rate decreases much faster ($\sim 8.5\%/cm$) for only the bottom source [2]. The reason for the slow decrease with the present thermal-neutron system is that the

thermal neutrons are being produced in the walls and the top lid, so the sample is being interrogated from all sides. Another reason for the slow decrease in the doubles rate versus height is that the efficiency for counting the IF reaction neutrons is higher as the sample moves away from the poly end plug that is an absorber for the neutrons. The higher efficiency gives a higher doubles rate and better correlation coupling to the ^{252}Cf background.

The doubles error was at the minimum when the sample was sitting on the bottom plug, so the calibration of the COE type samples will be performed with the can in the bottom position.

Electronics Set Up

The neutron data was collected using a JSR-15 shift register [5] and a laptop computer. The INCC software was used with the parameter setting listed in Table 1.

Table 1 INCC parameter values for the ANTECH AWCC at the calibration

Parameter	ANTECH AWCC	CI AWCC
High Voltage	1680 V	1680 V
Gate	128 μs	128 μs
Pre-delay	2.5 μs	2.5 μs
Efficiency	38.2%	38.2%
Deadtime a	0.28 μs	0.757 μs
Deadtime b	0.0196 μs	0.143 μs
multiplicity DT	70 ns	189 ns

Dead-Time Measurement

The dead time was measured using the 2 source method with the combined rates of $\sim 500,000$ cps. The singles dead time was slightly less than the doubles dead time, but because the doubles rate is the primary focus of the AWCC, we used a values that were dominated by the doubles coefficient. Note that we used $b = a^2/4$ and multi dead time = $a/4$.

6. MEASUREMENT PROCEDURES AND SAMPLE DESCRIPTION

The samples that were used for the optimization study included the U_3O_8 powder mixtures from NBL of the type being supplied to the COE. All the samples contained 230g of powder with enrichments varying from 0.31% to 93%. The sample cans were 7.6cm diameter and 8.6cm tall. Table 2 lists the sample contents.

Table 2 Calibration U_3O_8 sample description

U_3O_8 ID	U_3O_8 g	^{235}U % enrichment	^{235}U g
NBL-0075	230	93	181.09
NBL-0074	230	54	105.22
NBL-0073	230	20	38.98
0005-20.06	230	20.06	39.10
0006-52	230	52.56	102.40
446-071	230	4.46	8.69
295-072	230	2.95	5.75
194-071	230	1.94	3.78
En-0.71	230	0.71	1.38
En-0.31	230	0.31	0.60

The measurement procedure was to place the sample can on the HDPE bottom end plug and take a minimum of 30 cycles of 20s each for 600s total time. In some case much longer measurements were performed to better determine the statistical error. All errors in the tables have been normalized to 600s for comparison purposes. The singles and doubles backgrounds were measured with the blank (dummy) sample in the detector. The dummy sample had the same total mass as the calibration samples, but contained no fissile material. The room backgrounds were negligible because of the high source yield from the source FTC-CF-2494 ($1.422\text{E}+06$ n/s on 10/1/2015).

7. MEASUREMENT RESULTS

The U_3O_8 powder samples were measured sitting on the bottom HDPE end plug for at least 600s where the LEU samples were measured longer to obtain better statistical precision. The data was collected in 30s time bins so that the scatter in the data provided the statistical error estimates for each measurement.

Table 3 presents the calibration data and the statistical errors for a 600s measurement. The doubles background was determined to be 81700cps by a long overnight measurement using a dummy sample that contained no ^{235}U . The statistical error in the doubles background was negligible because of the long measurement time. Also, if the same source (after ^{252}Cf time decay) is used for future measurements of similar samples, then any uncertainty in the ^{252}Cf background cancels in the calibration curve parameters.

Table 3. The ANTECH AWCC measured results for U3O8 samples listed in Table 2.

U3O8 ID	235U g	Cf gross S dummy cps	Cf gross D dummy cps	Cf gross D cps	Cf net D	Cf 600s sigma cps	Cf 600s sigma %	Cf net S
NBL-0075	181.09	378600	81700	119253	37553	425.6	1.13	38954
NBL-0074	105.22	378600	81700	114304	32604	359	1.1	34037
NBL-0073	38.98	378600	81700	103677	21977	474.4	2.16	23770
NBL-0075	181.09	378600	81700	118550	36850	501	1.36	38900
NBL-0074	105.22	378600	81700	113838	32138	416.7	1.3	34159
NBL-0073	38.98	378600	81700	104030	22330	416.7	1.87	23687
0005-20.06	39.1	378600	81700	103526	21826	326	1.49	23742
0006-52	102.4	378600	81700	112867	31167	425.1	1.36	33941
446-071	8.69	378600	81700	89114	7414	440.7	5.94	7858
295-072	5.75	378600	81700	86966	5266	416.8	7.92	5962
194-071	3.78	378600	81700	85640	3940	340.5	8.64	3235
En-0.71	1.38	378600	81700	83709	2009	352.5	17.55	1167
En-0.31	0.60	378600	81700	82500	800	389.5	48.69	500

We observe in the Table 3 that the statistical error for a 600s measurement is ~ 1.1-1.8% for the mass range from 40-180 g ^{235}U . For mass values below ~ 20g, the error increases because of the low signal/background ratio. Note that the net doubles rate is almost the same magnitude as the net singles rate. This shows that the effective multiplicity is significantly higher than for a passive ^{252}Cf source in the sample position where the doubles/singles ratio is ~ 0.41. For a random source, the IF doubles/singles ratio is even smaller than for ^{252}Cf . The increase in the net doubles for the sample is a result of the time correlation between the ^{252}Cf background counts and the IF reactions.

Figure 6 shows the net doubles rate versus the ^{235}U mass both before and after subtracting the constant doubles background that was measured using the dummy sample. Fitting a 4th power polynomial through the data gives a zero mass intercept of 82461cps before the ^{252}Cf doubles background subtraction and 761cps after the background subtraction. This indicates that there is a fast-neutron IF component in the ^{238}U of about 761cps from the high energy ^{252}Cf neutrons. This component is only ~ 2-3% of the doubles rate for the higher mass samples, and it is built into the calibration function displayed on the graph.

The upper curve fitting function shown in Fig. 6

$$y = -7.47\text{E-}05x^4 + 0.04191x^3 - 8.5626x^2 + 821x + 82461$$

is the calibration function for the AWCC to be entered into the INCC software. The polynomial function is used to allow for an upward inflection in the AWCC calibration that is needed for future samples that have higher multiplication than the current samples.

Fig. 6 The doubles rate as a function of ²³⁵U mass before (top) and after (bottom) the subtraction of the ²⁵²Cf background.

The calibration function and coefficients are shown in Fig. 6 where the neutron source was COE FTC-CF-2494 with an emission rate of 1.422E+06 n/s (on 10/1/2015). To provide a realistic shape for the 4th order polynomial, a phantom data point was inserted off scale at 220g to force the curve to the desired shape from a physics standpoint. The polynomial functions shown in Fig. 6 should never be extrapolated beyond the actual calibration data range shown in the figure, because the extrapolated function goes very haywire.

8. COMPARISON OF AmLi NEUTRON SOURCE WITH ²⁵²Cf SOURCE INTERROGATION

For each of the calibration sample measurements using ²⁵²Cf, a similar measurement was made using the standard AmLi source MRC-95 for comparison purposes. The AmLi source was placed in the same position as the ²⁵²Cf source with the same thermal-neutron configuration of the AWCC. The same HV, gate and pre-delay were used for the AmLi interrogation. Thus, the only changes were the lower neutron energy spectrum and the random source interrogation. It has been shown in the past that for neutron doubles counting, the doubles error is independent of the interrogation source strength for the case where the accidental rate dominates the statistical error [6].

Table 4 presents the comparison of the ²⁵²Cf results with the AmLi results after normalizing the ²⁵²Cf source strength to the MRC-95 neutron **yield (32620 n/s)** at the time of the measurements (10/1/2015). The key observation is that the doubles rate from the normalized ²⁵²Cf source is ~

2.6 times higher per source neutron than for the AmLi source. The reason for the high net doubles rates for ^{252}Cf is that the IF reactions are time correlated with the background counts so the doubles tally in the coincidence gate gets the TCIF boost where **more than half of the net doubles originate from time correlated ^{252}Cf events**. Note that a singles background count that is time correlated with a singles neutron count from the IF in the 128us gate, results in a net doubles count for the signal. Figure 7 shows the net doubles rates after the normalization to the same source yield.

Fig. 7 Net doubles rate comparison for ^{252}Cf and AmLi after normalization to the same neutron interrogation strength of 32650n/s.

Table 4 Comparison of ^{252}Cf interrogation with AmLi random source interrogation

^{235}U g	AmLi	AmLi	AmLi 600s	Cf norm	Cf 600s	D Ratio
g	Net S	net D	sigma %	36250n/s	sigma %	Cf/AmLi
181.09	747	331.31	1.59	857.8	1.13	2.59
105.22	655	281.81	1.64	744.7	1.10	2.64
38.98	434	197.97	2.11	502.0	2.16	2.54
181.09	750	337.12	1.55	841.7	1.36	2.50
105.22	655	284.38	1.80	734.1	1.30	2.58
38.98	438	194.26	2.11	510.0	1.87	2.63
39.10	405	201.06	2.55	498.5	1.49	2.48
102.40	609	276.04	1.73	711.9	1.36	2.58
8.69	120	81.41	5.66	169.3	5.94	2.08
5.75	84	62.68	6.38	120.3	7.92	1.92
3.78	31	47.16	7.18	90.0	8.64	1.91
1.36	0.00	14.80	24.30	45.9	17.55	na
0.60	0.00	6.39	58.50	18.3	48.69	na

Another interesting result from Table 4 is that the statistical errors for the ^{252}Cf interrogation were about 30% lower than for the AmLi source interrogation in spite of the high ^{252}Cf doubles background where the signal/background ratio was $\sim 1/3$ for the ^{252}Cf source.

The statistical uncertainty for the correlated doubles would be very complex, because the measured counts do not follow simple counting statistical theory. Thus, we have used the repeat sample based method to actually measure the spread in the data for the doubles errors in Table 4.

Table 4 shows that the Cf/AmLi doubles ratio is increasing from 1.91 to 2.6 in going from LEU samples to HEU ($> 20\%$ enrichment). This is caused by the change in the correlated coupling coefficient between the ^{252}Cf source and the sample. When the ^{238}U fraction is much larger than the ^{235}U fraction (0.31 to 4.45%) for the 5 lowest enrichment samples in Table 3, some of the thermal neutrons that provide the IF in the sample are lost to parasitic absorption in the ^{238}U . For the HEU, the fractional loss to ^{238}U becomes negligible, and the ratio of the Cf/AmLi doubles is rather constant at ~ 2.6 for the AWCC.

9. SUMMARY

The optimization and calibration of the AWCC was performed for the ANTECH AWCC detector. The other two AWCCs (from Canberra), that are included in the COE project, were cross calibrated for efficiency and performance with the ANTECH detector. All 3 AWCC units have the same HDPE design and matched ^3He tubes (4 atm), and their operating parameters are the same except for the dead-time. The dead-time coefficients for the three AWCC systems are listed in Table 1. The dead-time difference was a result of the de-randomizing buffer in the

ANTECH junction box. The high neutron yields of the COE ^{252}Cf sources require a significant dead-time correction.

After the optimization of the AWCC for the ^{252}Cf source use, the doubles statistical error was about 1.2% for a 600s measurement of a 100g ^{235}U sample. This was about 30% lower than for the AmLi source interrogation with both sources in the thermal-neutron mode. The sensitivity limit of the AWCC using ^{252}Cf is $\sim 1\text{g }^{235}\text{U}$.

This report shows that the ^{252}Cf sources are viable replacements for the AmLi sources that are no longer available. The calibration function is a 4th order polynomial that is only valid within the calibration range 1-190 g of ^{235}U . When the unknown samples are measured with the same calibration function, any error related to the doubles background subtraction cancels in the measured ^{235}U mass result. The modification to the INCC software to support the use of the ^{252}Cf sources for the AWCCs is currently in progress [7].

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