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An Evaluation of a Neutron Time Correlated Interrogation Method for Measurement of Fissile Content in Research Reactor Spent Fuel Assemblies

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BACKGROUNND

There are many research reactors worldwide that have been, or are being converted from the use of high enrichment uranium (HEU) to low enrichment uranium (LEU, < 20% enriched). The verification of fissile content and initial enrichment of the spent fuel is needed for the effective safeguards of the fuel. The advanced experimental fuel counter (AEFC) was developed for the measurement of spent fuel rods and assemblies from research reactors for safeguards verification. This measurement system contains components for active neutron interrogation, passive neutron totals counting, neutron coincidence counting, and gross gamma-ray counting. This report presents the first application of the time correlated interrogation technique for the measurement of the ^{235}U content in research reactor spent fuel assemblies. The technique, called time correlated induced fission (TCIF), uses a ^{252}Cf neutron source to irradiate the fuel assembly, and the subsequent induced fission events in the fissile material are measured by coincidence counting. The doubles rates are enhanced by having the neutron trigger events from both the ^{252}Cf source and the induced fission neutrons in the same time gate in the coincidence analysis. The average neutrons per fission of the ^{252}Cf source is 3.76 and the induced fission neutrons for ^{235}U is 2.44, so the number of neutrons that are produced is higher than for random neutron interrogation. This high effective neutron number increases the multiplicity counting rates and reduces the statistical error. The background coincidence counts from the ^{252}Cf are reduced by the water in the sample cavity and the polyethylene surrounding the ^{3}He detector tubes. This method of active neutron interrogation has been applied to the measurement of spent research reactor (IRT) fuel assemblies. The advanced experimental fuel counter (AEFC) was used to compare the TCIF method with the typically used AmLi neutron interrogation source that emits neutrons that are random in time. The statistical uncertainty for the use of the random neutron source (AmLi) and the time correlated source (^{252}Cf) for spent fuel interrogations was evaluated.

1. INTRODUCTION

The AEFC was developed for the measurement of the ^{235}U content and enrichment properties of spent fuel elements from research reactors as well as rods from MAGNOX (**Magnox** is short for **Mag**nesium **n**on-**ox**idising clad fuel) reactors. This measurement system contains components for active neutron interrogation, passive neutron totals counting, neutron coincidence counting, and gross gamma-ray counting. The active assay mode uses two measurement methods: 1) neutron coincidence counting, and 2) total neutron singles counting. This report only address the active neutron interrogation analysis for singles and doubles rates. A more general documentation of the AEFC measurement activity is available elsewhere [1].

The AEFC was used for measurements of spent fuel produced from operation of the WWR-SM research reactor at the Institute of Nuclear Physics (INP) in Tashkent, Uzbekistan on Oct 4-13, 2011, and again on Sept 15-20, 2014. More than twenty fuel assemblies were measured in 2011 of the IRT-3M design [2], as well as twenty-two fuel assemblies that were measured in 2014. The assemblies covered a wide range of initial enrichment, burnup and cooling times. This report

presents the results of the active mode neutron interrogation using the AEFC for spent fuel at the research reactor at INP for both a random neutron source (AmLi) and a time correlated source (^{252}Cf). The measurements were the first application of the AEFC using the new ^{252}Cf based TCIF measurement method [3-4] to research reactor fuel.

The AmLi neutron sources that have been used for the AEFC and other active neutron interrogation instruments are no longer commercially available, and alternative neutron sources are needed. The TCIF method makes use of spontaneous fission neutron sources such as ^{252}Cf 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 AEFC systems used an AmLi source of ~ 1.0 Ci to provide a neutron yield of ~ 4.5×10^4 n/s; whereas, a 100μCi ^{252}Cf source yields 4.4×10^5 n/s. Thus, the Ci level is reduced by roughly four orders of magnitude by using the ^{252}Cf source. In addition to this improvement in radiation level and initial cost, the use of the ^{252}Cf source can reduce the required measurement time by a factor of ~ 2-3 to reach the same assay precision as for the AmLi source. This improvement is primarily due to the improved signal/background ratio for the ^{252}Cf source. The data to support these performance improvements is presented in this report.

1. TIME CORRELATED INTERROGATION CONCEPT

For the TCIF method, the fuel assembly is interrogated with neutrons from the ^{252}Cf source; however, the measured doubles rates are enhanced because the IF reactions 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. Also, there is a significant probability for more than one induced fission reaction from a single spontaneous fission reaction. 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 a random interrogation source such as AmLi. This high effective ν significantly increases the multiplicity counting rates and reduces the statistical error. The background coincidence counts from the ^{252}Cf are reduced by the water and polyethylene neutron shielding between the source and the detector tubes. There is still a residual background from the ^{252}Cf source, and this background is proportional to the source strength. However, this ^{252}Cf source background is much smaller than the induced signal, and it can be measured once for all assemblies for a measurement campaign that uses the same source.

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 80 μs for the AEFC, but also the die-away time of the interrogation IF that is about 50 μs. Thus, to have our gate interval include both SF and correlated IF, we have increased the gate to more than 100 μs. However, the statistical error is a function of the accidental counts (A) pileup [5-6], and a longer gate will make the A related error larger. An analysis of the data indicated that a gate length of 128 us was near optimum for the reduction of the statistical counting error in the doubles rates.

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

1. Neutrons from the ^{252}Cf source (doubles and singles)
2. Passive neutrons from the spent fuel assembly (doubles and singles)

A separate passive neutron measurement is used to determine the background from the spent fuel assembly, and the ^{252}Cf source background was measured separately without a fuel assembly in the detector. An inert fuel assembly with no fissile content would provide a better estimate for the ^{252}Cf background but such an assembly was not available. The neutron background from the other spent fuel assemblies in the storage pool was negligible. The ^{252}Cf background was measured overnight so that its statistical error was very small. The ^{252}Cf source background was only about 10% of the induced doubles signal, and the sum of both backgrounds (^{252}Cf and the fuel assembly) represent only about 20% of the IF coincidence signal.

3. SYSTEM DESCRIPTION

The AEFC consists of a cylindrical polyethylene moderator, lead shielding, six ^{3}He tube neutron detectors, and an ion chamber surrounded by a stainless steel body that is water tight. The fuel element through-hole diameter is 117 mm. **Error! Reference source not found.** provides a photograph of the complete system together with the internal components where the ^{3}He tubes are surrounded by Pb shielding for gamma-ray reduction. The shielding reduces the gamma dose on the tubes to less than 100 R/h (1 Sv/h) for the highest burn-up fuel assembly. The six ^{3}He tubes have gas pressure of 4 atmospheres and each tube was connected to a PDT-10A amplifier. The tubes are designed to measure neutrons without gamma interference for fuel assembly surface dose levels up to 10^4 R/h (100 Sv/h) [7]. Higher dose levels can be measured by reducing the high voltage (HV) bias on the ^{3}He tubes and operating at a reduced efficiency. Each of the ^{3}He tubes is connected to a PDT-10A amplifier that is gain adjusted to reject the gamma pileup activity. The neutron data was collected using a JSR-15 shift register [8] and a laptop computer.

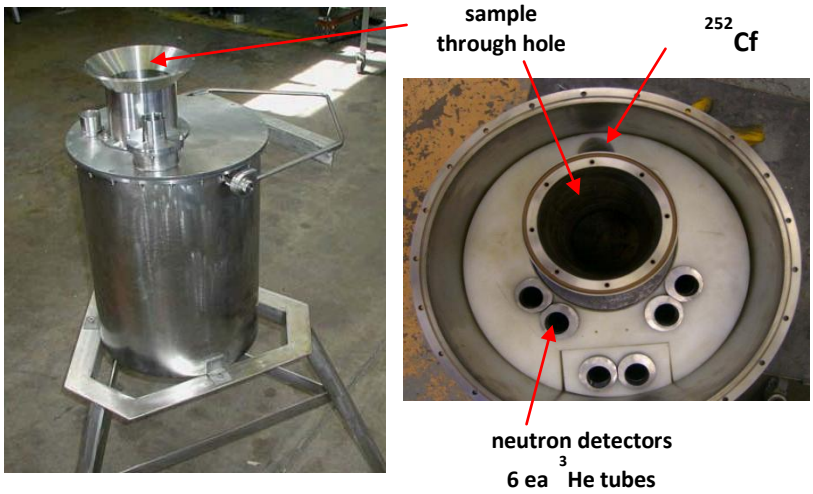

Fig. 1. Diagram of the AEFC showing the six 3He tubs, the Pb shielding and the HDPE moderator.

4. FUEL ASSEMBLY DESCRIPTION

The spent fuel elements for the INP reactor have both rod-type fuel and concentric box shells design. The fuel is uranium aluminum alloy (UAl_x) clad in aluminum for the older IRT type fuel, and the newer 19.8% enriched LEU fuel has a uranium molybdenum alloy to reach a higher density [2]. The type of fuel alloy affects the passive alpha reaction neutron yield from the alloy but does not significantly impact the passive doubles rate and the active neutron measurement for the ^{235}U content.

The specifications for the spent fuel elements that were measured during the 2011 exercise are listed in **Error! Reference source not found.** [9]. The residual ^{235}U mass was derived from the facility records for the initial ^{235}U mass reduced by the burnup (BU) record.

Table 1: IRT-3M type fuel specifications for the reactor at INP 2011 campaign [9].

Item ID	Initial Enrich. [%]	BU [%]	Remaining 235U [g]	Fuel Type
231	36	71.94	88.33	Box
28	36	29.1	216.95	Box
359	36	60.4	123.08	Box
141	36	60.7	119.2	Box
232	36	71	91.32	Box
215	36	31.57	214.94	Box
84	36	41.1	179.82	Box
186	36	62.22	114.55	Box
140	36	59.9	122.51	Box

131	36	56.1	133.63	Box
143	36	59.3	125.4	Box
183	36	60.02	120.02	Box
19	36	39.49	183.41	Box
228	36	70.9	91.37	Box
142	36	62.6	114.71	Box
126	36	58.42	125.41	Box

The fuel assemblies that were measured in 2011 included several initial enrichments, but only the 36% enrichments are analyzed in this paper. However, for the 2014 measurements, only 10% and 19.8% enrichment assemblies were available. Table 2 list the IRT-4M type fuel assemblies that are included in the active neutron measurements in 2014 that are included in this paper. The BU for the EK-10 assemblies was not known and the average value of 29% is listed in Table 2.

Table 2: INP reactor declared spent fuel specifications (2014 campaign).

Fuel Type	**Assembly ID**	**Initial Enrich. [wt%]**	**Burnup [%]**	**Residual ^{235}U Mass [g]**	**Cooling Time [years]**
IRT-4M	430	19.75	60.96	99.9	2
IRT-4M	415	19.75	61.46	102.4	3
IRT-4M	428	19.75	55.27	113.93	3
IRT-4M	433	19.75	56.59	111.04	2
IRT-4M	414	19.75	59.93	105.02	3
IRT-4M	398	19.75	56.98	114.8	2
IRT-4M	413	19.75	57.01	114.74	3
IRT-4M	418	19.75	57.42	112.84	3
IRT-4M	411	19.75	59.38	107.6	3
IRT-4M	417	19.75	56.78	115.22	3
IRT-4M	04M	19.75	66.5	99.76	12
IRT-4M	07A	19.75	60	105.8	12
EK-10	10A	10	~ 29	TBD	42
EK-10	10B	10	~ 29	TBD	42
EK-10	10C	10	~ 29	TBD	42
EK-10	10D	10	~ 29	TBD	42
EK-10	10F	10	~ 29	TBD	42
EK-10	10H	10	~ 29	TBD	42
EK-10	10I	10	~ 29	TBD	42
EK-10	10J	10	~ 29	TBD	42
EK-10	10K	10	~ 29	TBD	42

5. MEASUREMENT PROCEDURES

There were different measurement procedures for the 2011 exercise compared to the 2014 measurements. For the 2011 data, the center of the fuel assembly was positioned in approximately the center of the neutron detectors and the assembly was measured for 600 s in the passive mode followed by 600 s in the active interrogation mode using 30 s data cycles. After the passive count, the AmLi neutron interrogation source was inserted into the AEFC through a PVC tube to obtain the active measurement.

Fig. *1* shows the AEFC system being deployed at the INP reactor site in 2011 where the AEFC is setting on top of the spent fuel storage rack. The spent fuel assemblies in the storage rack directly below the AEFC did not change the measured neutron background level because the AEFC stand was ~ 510 mm tall with water shielding between the detector and the storage fuel. The white PVC tube is shown connecting the detector to the surface of the pool. The interrogation source was connected to a Teleflex cable for insertion and removal through the PVC tube by hand. The interrogation neutron source holders for both the AmLi and the ^{252}Cf sources are shown in Fig. 3. The AmLi source holder is made of tungsten to reduce the gamma dose; whereas, the ^{252}Cf source holder is made of HDPE for neutron moderation.

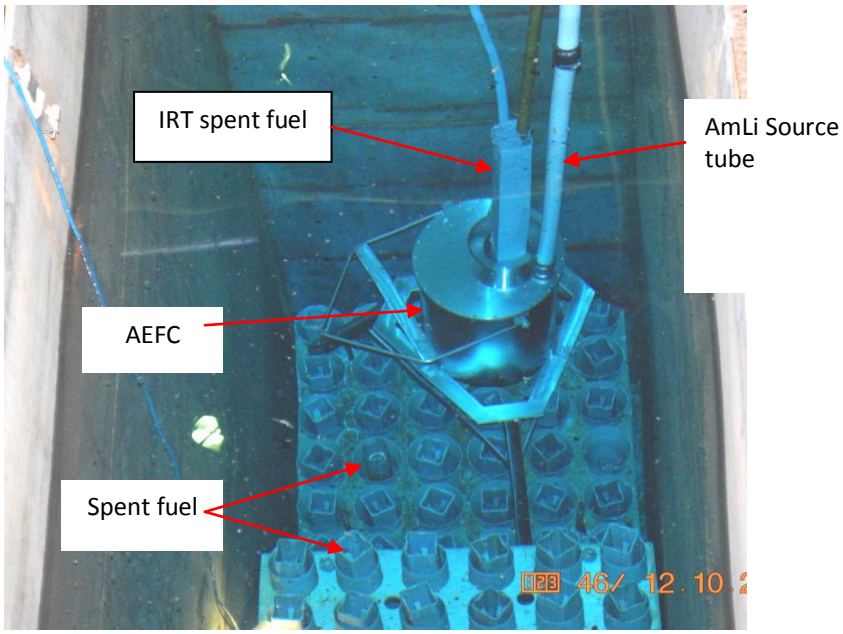

Fig. 1. Photograph of the deployed AEFC on the support stand on top of the spent fuel rack at the WWR-SM reactor site during the 2011 measurement campaign [9]

Fig. 3. Photograph of the tungsten AmLi source holder (left) and the ^{252}Cf HDPE source holder (right), beside the Teleflex cable-lid for transferring the sources into the AEFC in 2014.

For the 2011 measurements, there was considerable uncertainty in the sample vertical position that was based on finding the maximum in the passive neutron counting rate. Thus, for the 2014 measurements, a new 3 position measurement procedure was implemented. The new fixture had 3 measurement positions that were determined by a polyethylene fixture that was attached to the fuel assembly handling pole. The fixture had steps with a pitch of 25 cm. For the data collection, the fixture rested on a platform above the water pool and in line with the AEFC sample hole.

The primary purpose for the 3 position measurement was to provide a more uniform coverage of the residual ^{235}U mass in the spent fuel. The center position for the fuel assembly has a significant reduction in the ^{235}U concentration per unit length compared with the ends, and this makes the 3 position scan necessary to include the ^{235}U mass near the ends of the assembly. Note that the passive neutron background from the spent fuel assembly has the maximum neutron yield from the center position because of the higher plutonium buildup in the central region.

The AEFC should be operated at a HV that is low enough so that the gamma-ray pileup does not interfere with the neutron counting rate. The lead shielding for the AEFC was designed for operation at all burnup levels and all cooling times that are over ~ 2 years. To establish that there was no gamma interference in the neutron counts, we measured a HV plateau curve for the fuel assembly with the highest gamma activity (IRT-4M-430) and compared the plateau curve with an assembly with low burnup and a longer cooling time (2.2 years). Figure 4 shows the measured plateau curves from the 2011 exercise for the maximum gamma dose assembly (SFA359) compared with a low burnup assembly (SFA215). The high gamma dose was primarily caused by the short cooling time (0.54 years) that was less than the recommended 2 year cooling time.

We operated the AEFC at a HV of 1620 V to be below the gamma pileup threshold for all of the assemblies. The MTR calibration [11] HV was 1680 V, so our efficiency for the spent fuel measurements in 2011 was ~ 5% below the calibration value at 1680V. This efficiency correction is needed when comparing calibration data for different AEFC applications.

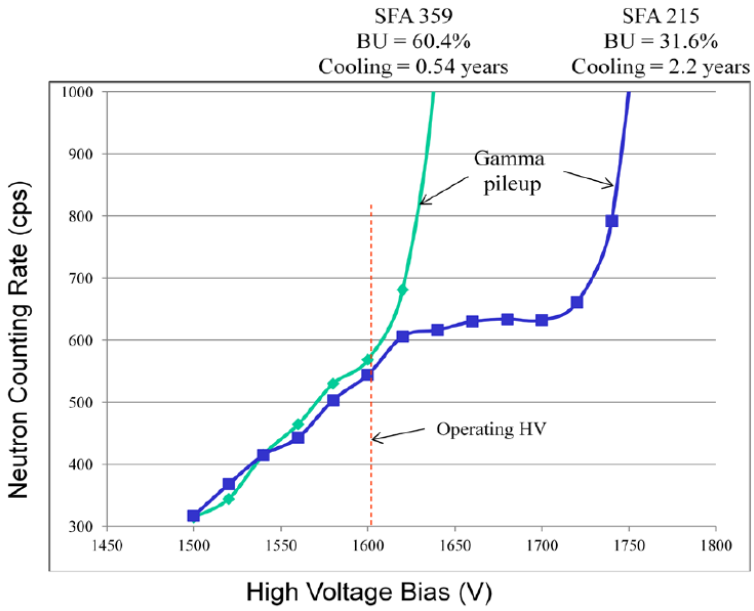

Fig. 4. High voltage plateau curves for the maximum gamma dose spent fuel assembly (SFA359) compared with a low BU fuel assembly (SFA215) in the 2011 measurements [9].

6. MEASUREMENT RESULTS

For the 2014 measurements, the fuel assemblies were measured using the 3 position fixture to cover the entire assembly active length. Each assembly was measured for 300 s passive and 300 s active at each position for a total time 30 min per assembly. The doubles mode pre-delay was set at 4.5 us and the doubles gate was set at 128 us. The data was collected in 30 s time bins so that the scatter in the data provided the statistical error estimates for each measurement. The average for the rates from the 3 positions was used for the analysis to improve the counting rate error and to cover the full active length of the assemblies. The average counting rates for the 3 positions are listed in Table 3. The doubles error corresponds to the data scatter for the 30 cycles of 30 s each (900 s) of data used to obtain the average rates. The errors related to the ^{252}Cf background subtraction (35.8 cps) and the fuel assembly passive background subtraction (~ 25 cps) are included in the error calculation. Because the backgrounds were only a small fraction of the doubles signal level, the resulting contributions to the total error were small.

Table 3: Net singles and net doubles rates for the ^{252}Cf interrogation source (2014 data).

Fuel Type	Assembly ID	Net S, Average [cps]	Net D Average [cps]	Sigma Doubles [cps]	Sigma Doubles [%]
IRT-4M	430	3334	214.4	7.02	3.28
IRT-4M	415	3386	220.2	4.86	2.2
IRT-4M	428	3735	257.9	6.26	2.43
IRT-4M	433	3751	241.8	6.07	2.51
IRT-4M	414	3616	236.9	5.35	2.26
IRT-4M	398	3468	250.5	8.79	3.51
IRT-4M	413	3771	249	5.28	2.12
IRT-4M	418	3880	258.4	5.72	2.22
IRT-4M	411	3529	239.8	5.29	2.2
IRT-4M	417	3522	249.2	7.92	3.18
IRT-4M	04M	3231	195.8	6.33	3.23
IRT-4M	07A	3301	217.1	4.87	2.27
EK-10	10A	2476	138.7	4.38	3.16
EK-10	10B	2264	122.9	3.2	2.61
EK-10	10C	772	32.2	2.73	8.48
EK-10	10D	3211	178.7	6.29	3.52
EK-10	10F	2616	148.4	5.35	3.6
EK-10	10H	768	22.3	1.49	6.71
EK-10	10I	2522	143.5	4.46	3.11
EK-10	10J	2555	137.3	4.61	3.36
EK-10	10K	1665	84.2	4.85	5.76

Figure 5 displays the average passive and net active singles and doubles results for the fuel assemblies listed in Table 3. The doubles rates are plotted versus the singles rates for a data constancy check. Also, for the passive data (left), the doubles increase with burnup faster than the singles because of the buildup of 240Pu for the higher burnup assemblies. The data shows that the passive doubles background rate is only about 20% of the net active signal for the doubles, and about 60% for the net singles rates. The ^{252}Cf source backgrounds of 35.8cps for the doubles and 5650cps for the singles has been subtracted from the measured data. The statistical error for the net active data is about the size of the data symbols. Note that in Fig. 5 (right), that the net active doubles increases faster with the ^{235}U mass than for the singles rate because the doubles increase with multiplication faster than singles. It is of interest that the highest data point for the passive data (IRT-4M-04) is the lowest data point for active interrogation of the IRT-4M assemblies because much of the ^{235}U mass has been burned out for the high burnup assembly. Also, the active doubles for the high burnup assemblies is reduced because of thermal-neutron absorption in the relatively high fission product absorbers. Also, because of the high burnup in assembly 04M, the thermal-neutron absorber ^{240}Pu is increased relative to the other assemblies.

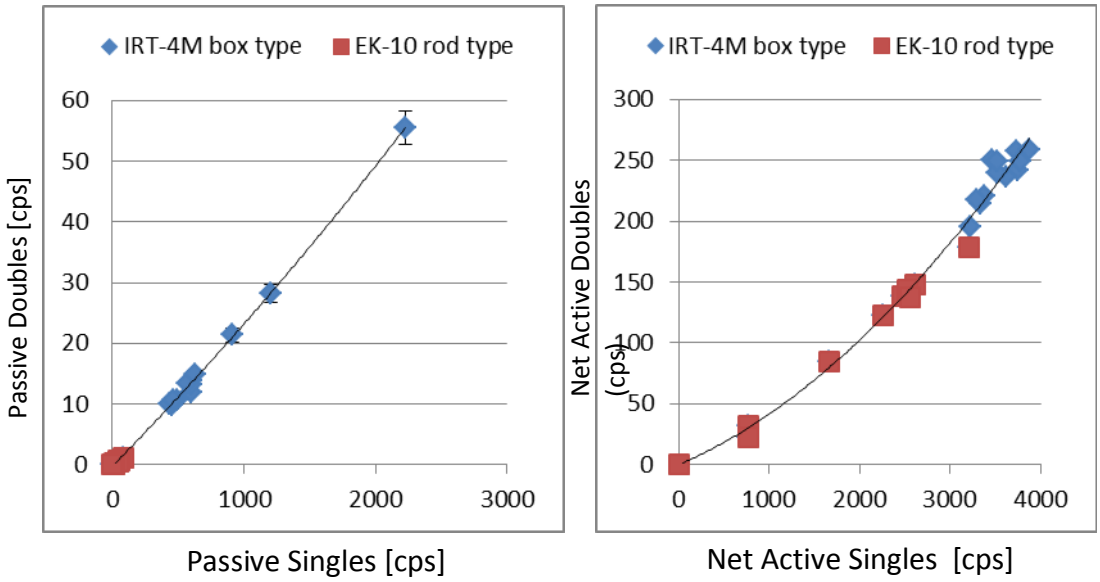

Figure 5. Passive doubles versus passive singles (left), and net active doubles versus net active singles (right) showing the doubles enhancement for the neutron interrogation (2014). Note that the burnup order of the assemblies has reversed in the two plots.

Figure 6 shows the relationship between the net active doubles and active singles rates versus the declared residual ^{235}U mass in the IRT 4M type fuel assemblies. We see that the doubles rate is increasing at a faster rate than the fissile mass because of the multiplication in the doubles rates.

The data from Fig.6 was fit with second order polynomials for both the singles and the doubles rates. The high mass assembly IRT-4M-04M (the green doubles data point) was removed from the trend-line fit because it has a higher initial ^{235}U and ^{238}U mass than the other IRT-4M assemblies.

Both of these curves (singles and doubles) can be used as calibration functions to measure the residual ^{235}U mass in the EK-10 fuel assemblies where the facility tag values were not available. However, because of the different assembly geometries (rods vs boxes), future MCNP simulations will be required to accurately connect the calibration curves for the different types of fuel assemblies.

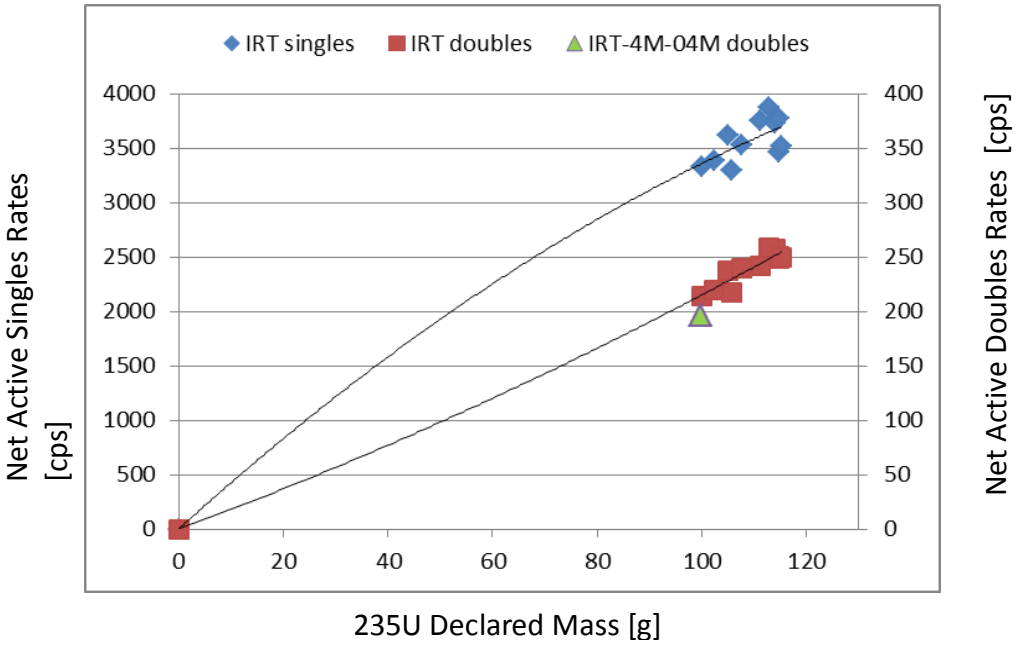

Fig. 6. Net active doubles and singles rates versus the declared 235U residual mass in the IRT-4M assemblies.

The net active mode doubles and singles rates for the EK-10 fuel assemblies are shown in Fig. 7 with a second order polynomial trend line through the data where the symbol for the normal EK-10 assemblies that contained 16 fuel rods. However, 3 of the assemblies had fewer rods and 1 had more rods (22), and these data points fall slightly below the 16 rod assemblies. The polynomial curve through the data was fit to only standard 16 rod configuration.

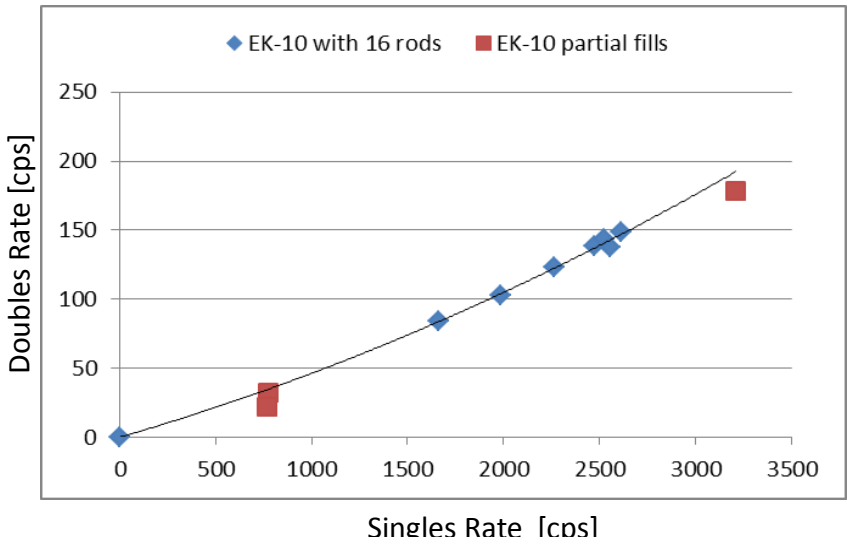

Fig. 7. Net active doubles rates versus active singles rates for the EK-10 assemblies.

The measured doubles and singles rates for the EK-10 assemblies were fit to the IRT-4M element curves shown in Fig. 6 to calculate the residual ^{235}U mass for each EK-10 assembly. These results are shown in Table 4 for both the doubles rates and singles rates. Note that the use of the IRT-4M data as a calibration for the EK-10 rod geometry makes the assumption that the neutron self-shielding and multiplication is the same for both the rod geometry as for the concentric box geometry. This assumption needs to be checked with future MCNP code simulations.

In principle, both the doubles and singles calibrations should provide the same result within the accuracy of the measurements. Figure 8 shows a graph of the ^{235}U mass as calculated by the two different calibration curves showing that the 16 rod EK-10 assemblies have approximately the same mass from both the doubles and singles calibrations. The three outlier data points (orange) could be caused by the reduced doubles multiplication for the modified fuel assemblies.

Table 4: Calculated ^{235}U Mass Values for EK-10 Assemblies Based on IRT-4M Calibration Curves.

EK-10 ID	Net D (cps)	Net S (cps)	^{235}U via S (g)	^{235}U via D (g)	^{235}U Av. (g)
10A	138.7	2476.1	75.8	75.1	75.4
10B	122.9	2263.9	69.3	68.1	68.7
10C	32.2	772.2	18.2[a]	15.6[a]	16.9[a]
10D	178.7	3211.4	98.3	90.9	94.6
10F	148.4	2615.6	80.0	79.1	79.6
10H	22.3	768.1	23.5	14.2	18.8
10I	143.5	2522.3	77.2	77.1	77.2
10J	137.3	2555.1	78.2	74.5	76.3
10K	84.2	1664.6	50.9	49.4	50.1

a) A 30% reduction correction was made for assembly EK-10C because the active data indicated that fuel mass was concentrated in the bottom of the fuel can.

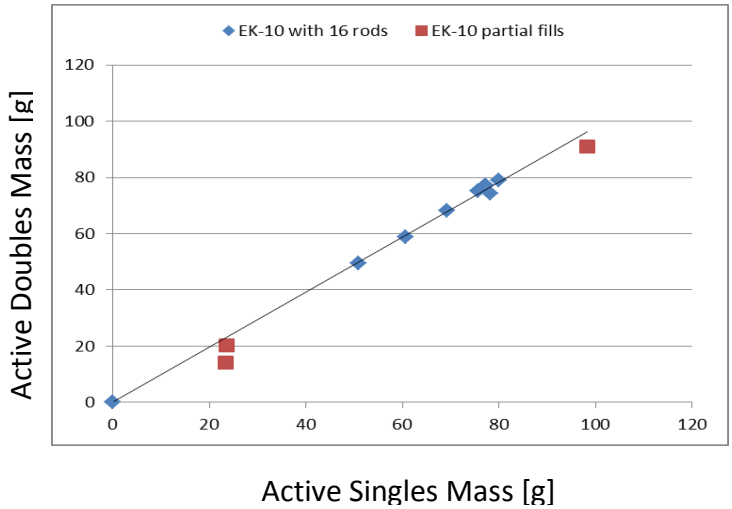

Fig. 8. Net active doubles calculated ^{235}U mass versus singles rates ^{235}U mass for the EK-10 assemblies

We have used the average values of the two different calibration results to provide the best estimate of the mass loadings in the EK-10 assemblies. The active assay results and error estimates for all of the IRT-4M assemblies measured in 2014 are shown in Fig. 9 where the statistical errors were determined via the 3x30 repeat cycles of 30s each. The displayed error bars are at 3.5% to include both the measured doubles statistical errors (~ 2.5%), and potential positioning uncertainties in the measurements.

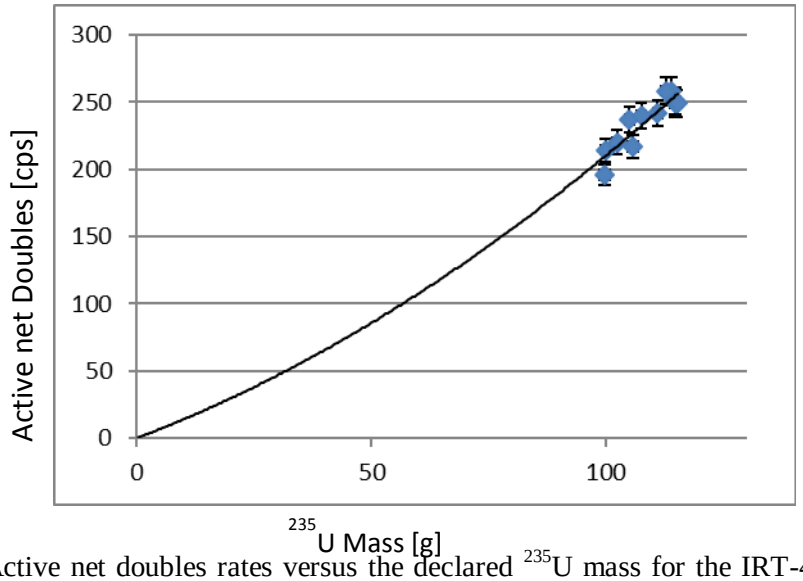

Fig. 9. Active net doubles rates versus the declared ^{235}U mass for the IRT-4M assemblies showing the estimated 1 sigma errors in the measurements and the second order polynomial trend line.

Table 5: Comparison of the scatter in the measured ^{235}U mass with the declared mass for IRT-4M assemblies (2014 data).

IRT ID	^{235}U Tag [g]	^{235}U [g] via S	S Diff. [%]	^{235}U [g] via D	D Diff. [%]
430	99.9	102.21	-2.31	101.43	-1.53
415	102.4	103.82	-1.39	103.41	-0.99
428	113.93	114.51	-0.51	115.83	-1.66
433	111.04	115.00	-3.57	110.63	0.37
414	105.02	110.87	-5.57	109.02	-3.81
398	114.8	106.34	7.37	113.45	1.18
413	114.74	115.63	-0.78	112.98	1.54
418	112.84	118.97	-5.43	115.99	-2.79
411	107.6	108.21	-0.57	109.97	-2.2
417	115.22	107.99	6.27	113.02	1.91
04M	99.76	99.07	0.69	94.91	4.86
07A	105.8	101.21	4.34	102.35	3.26

%RSD	4.2	2.61

The statistical counting error for the net singles rates is smaller than for the net doubles rates, yet Table 5 shows that the doubles scatter versus the declared mass calibration is only 2.61% compared with 4.2% for the singles mode. One reason for the smaller error in the doubles mode is the shape of the calibration curves. Figure 6 illustrates that the calibration curve is steeper for the doubles rates, whereas, the singles curve is slightly concave. Thus, the doubles statistical error is slightly reduced in the transfer to the ^{235}U mass.

7. COMPARISON OF AmLi NEUTRON SOURCE INTERROGATION WITH ^{252}Cf SOURCE INTERROGATION

The AEFC was originally designed to use and AmLi neutron source for the active assay mode. However, we have substituted a ^{252}Cf source (~ 7.37×10^5 n/s) for the AmLi neutron source (~ $4.6\text{x}10^4$ n/s) to improve the signal/background ratio, and to reduce the counting statistical error. We have compared data and errors using an AmLi source from the 2011 measurement campaign with the ^{252}Cf source in 2014. Fortunately, one of the 2011 fuel assemblies (126) was measured using both AmLi and ^{252}Cf for a direct comparison of the two types of sources during the 2011 measurements. Table 6 lists results from the 2011 active assay measurements for the IRT box type assemblies. We see that the RSD differences from the operator tag values was 13.2% for the doubles and 15.9% for the singles measurements for the 2011 data.

Table 6: Measured IRT Assemblies Using AmLi Neutron Interrogation in 2011.

IRT ID	BU [%]	Tag ^{235}U [g]	D mass [g]	S mass [g]	D diff. [%]	S diff. [%]
231	71.9	88.33	96.69	68.96	-9.46	21.93
28	29.1	216.95	–	207.86	–	4.19
359	60.4	123.08	122.92	119.29	0.13	3.08
141	60.7	119.2	90.61	107.48	23.98	9.83
232	71	91.32	104.46	129.21	-14.39	-41.49
215	31.6	214.94	180.4	200.41	16.07	6.76
84	41.1	179.82	154.29	158.25	14.2	11.99
186	62.2	114.55	122.87	127.48	-7.26	-11.29
140	59.9	122.51	93.24	115.02	23.89	6.11
131	56.1	133.63	131.93	124.19	1.28	7.07
143	59.3	125.4	129.72	120.48	-3.45	3.29
183	60	120.02	114.72	119.56	4.41	0.38
142	62.6	114.71	91.95	83.95	19.84	26.81
126	58.4	125.41	132.69	127.01	-5.81	-1.28
%RSD					13.2	15.9

Figure 10 shows the active mode net singles rates for the original AEFC calibration at LANL [11] using an AmLi neutron interrogation source with a fresh MTR type plate fuel assembly

(left), and the data taken for spent IRT fuels assemblies in the 2011 campaign (right). Both data sets show the nonlinear shape caused by the thermal-neutron absorption (self-shielding) in the fuel. Also, the IRT spent fuel assemblies (2011) showed considerable scatter compared to the declared values because of both counting statistics and sample positioning uncertainties.

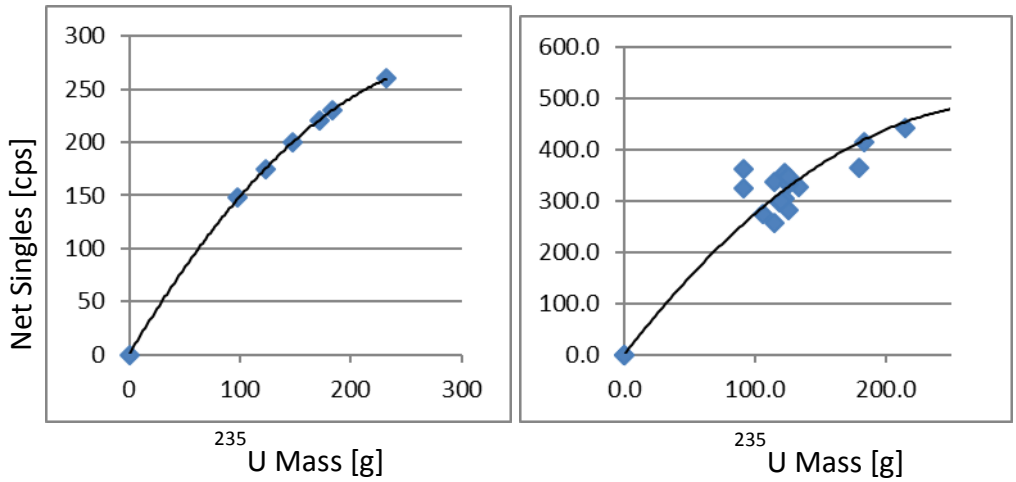

Fig. 10. Active mode singles rate response curves for the MTR fuel calibration and the IRT fuel assemblies using an AmLi neutron interrogation source in 2011.

Figure 11 shows the AmLi net doubles data from 2011 that can be compared with the ^{252}Cf data from 2014 shown in Fig. 9 where the data scatter is much less for the ^{252}Cf source interrogation. The average deviation from the tag values for the AmLi source was 13.2% compared with 2.6% (Table 5) for the ^{252}Cf interrogation results from 2014. It should be noted that the 2011 fuel assemblies were HEU (36% enriched) compared with LEU (19.8%) for the 2014 assemblies. Part of the scatter in the 2011 data shown in Fig. 11 was caused by the positioning uncertainty that was removed by the 3 position scanning procedure in 2014.

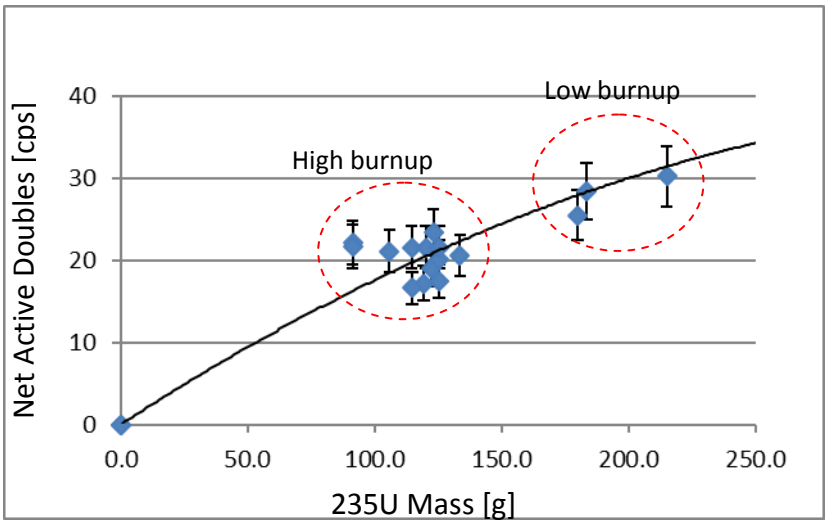

Fig. 11. Active mode doubles rate versus the declared mass for IRT fuel assemblies (2011 data)

It should be noted that all of the fuel assemblies in Fig. 11 were measured using the AmLi source. Also, they all had 36% initial enrichment (~ 850g ^{238}U).

7.1 Direct Comparison of AmLi and ^{252}Cf for Spent Fuel Assembly 126 in 2011

During the 2011 measurement campaign, a single IRT-3M fuel assembly (126) was measured with both the AmLi neutron source and a ^{252}Cf source that was on loan from INP for the day. This provided a direct comparison of the statistical error between the two types of sources where all of the sample and detector parameters remained the same. The sample position was invariant, and the efficiency, gate setting (100 us), pre-delay (4.5 us), high voltage bias, and die-away time remained the same for both neutron sources.

The statistical error was determined from the repeat cycles for the singles and doubles rates for both neutron sources. The measured rates are given in Table 7 for the two neutron sources that were used for the same sample. We see that the statistical error for the net doubles is ~ 250% smaller for the ^{252}Cf source than the AmLi source. Most of this improvement comes from the higher signal/passive background ratio for the ^{252}Cf source. The high counting rate for the ^{252}Cf source makes the passive neutron background less than 10% of the induced doubles rates. The ^{252}Cf time correlated induced fissions in the sample also helped to reduce the doubles statistical error because of the time- correlated trigger events from the ^{252}Cf.

Table 7: Measured data for IRT-3M assembly (126) using both AmLi (N531) and ^{252}Cf neutron interrogation in 2011.

Source	Pass S [cps]	Pass D [cps]	Active S [cps]	Active D [cps]	net act. D [cps]	A [cps]	% error D [600s]
AmLi+126	1308	22.84	1839	36.04	12.9	338	6.17
^{252}Cf+126	1308	22.84	19048	408.6	330.4	36283	2.36
^{252}Cf bkg	n/a	n/a	10832	55.28	0		
AmLi bkg	n/a	n/a	203.3	**0**	**0**		

The statistical error for the ^{252}Cf doubles of 2.36% is consistent with the subsequent ^{252}Cf doubles errors of ~ 2-3% in 2014 (Table 3). The observed 260% improvement in the statistical error led to the follow-up measurements in 2014 that used a ^{252}Cf source in place of the AmLi source.

8. COMPARISON OF ^{252}Cf WITH AmLi USING A FRESH MTR FUEL ASSEMBLY (2015 data)

To better understand the comparison of ^{252}Cf sources with AmLi sources, a set of measurements using both type sources were performed at LANL for a fresh MTR type assembly in 2016 [Jay 13]. The measurements were in a water tank and the fuel assembly was positioned on the centerline of the ^{3}He tubes in the AEFC. The assembly had a ^{235}U fuel mass of 231.7 g in the standard MTR parallel plate type geometry. Because the assembly had never been irradiated, the

passive neutron background was negligible. The JSR-15 coincidence gate was set at 128 us with a 4.5 us pre-delay.

The results of the measurements showed that the AmLi source induced 16% more IF per source neutron than ^{252}Cf because of the lower average neutron energy for the alpha-Li reaction. However, the doubles rate per source neutron was 23% higher for ^{252}Cf because of the TCIF boost. For repeat measurement cycles of 30x20s (600 s), the statistical doubles error for the ^{252}Cf source was 20% smaller than for the random neutron AmLi source. The doubles/singles ratio for ^{252}Cf was 1.46 times higher than for the AmLi source showing the TCIF doubles boost. For these measurements, both sources had approximately the same neutron emission rate that was ~ $4.0x10^4$ n/s. Note that for low background conditions and high singles rates, the doubles error is relatively independent of the neutron counting rate. These measurements showed that the TCIF boost for a ^{252}Cf source improved the statistical error in the spent fuel measurements by ~ 20% independent of the improvement in the signal/passive background ratio for the ^{252}Cf.

A separate more complete calibration paper by Joshi has been prepared to provide more information related to the fresh MTR fuel assembly calibration measurements [13].

9. SUMMARY

This report compares the relative error when using a ^{252}Cf source compared with an AmLi neutron source for the interrogation of spent fuel assemblies. The time correlated interrogation method (TCIF) for the active neutron measurement of fissile material in spent IRT type fuel assemblies is described for two field tests of the AEFC. The method makes use of the time-correlated neutrons from a spontaneous fission source such as ^{252}Cf to induce fission reactions in the fissile material including ^{235}U and ^{239}Pu. The IF reactions are time correlated with the spontaneous fission source neutrons during the time gate for measuring the correlated neutrons, and this increases the measured neutron rate in the doubles gate.

The spent IRT fuel assemblies contain a small quantity of ^{239}Pu, and the AEFC active neutron measurement includes all fissile components. Published estimates [12] of the ^{239}Pu/^{235}U ratios for spent IRT fuel indicate that the typical mass of ^{239}Pu would be in the range of 3-8 g depending on the burnup. Future burnup calculations will be performed to better quantify the ^{239}Pu fraction in the IRT spent fuel. For the present report, we have included the small mass of Pu fissile material as a component of the ^{235}U mass in the figures.

The advantages of the ^{252}Cf source compared with the AmLi neutron source follow:

- A reduction in the doubles counting statistical error.
- An improvement in the active signal/background ratio.
  - The reduction in the 2011 measurement error for the ^{235}U mass from ~13% to ~ 2-3 %, because higher signal/background ratio and the better positioning from the polyethylene fixture.
- The ^{252}Cf sources are commercially available, and the radioactive isotope inventory for the ^{252}Cf interrogation source is reduced by four orders of magnitude compared to AmLi providing less safety concerns.

Because of the improvement in the statistical error, for future use of the AEFC, we could shorten the measurement times by a factor of ~ 2-3 and still obtain statistical errors that are less than 5% for the active mode measurement.

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