ABSOLUTE MEASUREMENTS OF WASTE USING A NEUTRON MULTIPLICITY DRUM MONITOR

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ABSTRACT

Neutron Multiplicity Counting (Neutron Triple Correlation Analysis) is increasingly employed in the measurement of plutonium containing waste. The technique brings the potential for both improved measurement accuracy and the ability to gain sample matrix information as part of the waste assay measurement. Neutron coincidence counting or neutron pair correlation using shift register electronics, allows one to determine the Totals and the Reals count rate from plutonium. In contrast, neutron multiplicity counting allows the determination of three parameters including the correlated triples rate. The technique employs the point source interpretation model which allows one to determine three of the four unknown assay parameters by measuring the Totals count rate, the Reals count rate (correlated pairs) and the correlated Triples rate. Thus, where one of the unknown assay parameters can be established or is known, then absolute measurements of waste are possible. The present paper describes the collaborative development of a multiplicity drum monitor for the measurement of plutonium containing waste in 200 litre drums. The device can be used with coincidence electronics although it is designed specifically for neutron multiplicity counting using the ANTECH neutron Time Correlation Analyser. Data is presented in this paper from measurements on the prototype drum monitor developed at the Joint Research Centre and on a recent production instrument. The measurements include both Pu and Cf252 samples as well as Plutonium measurements for an empty and a concrete drum containing a Pu sample.

INTRODUCTION

Plutonium present in radioactive waste, generates neutrons from spontaneous fission events, from (alpha,n) reactions and from induced fission events caused by primary neutrons. Neutron pair correlation (coincidence counting) and neutron triple correlation (neutron multiplicity counting) provide a means of determining the spontaneous fission rate F_S of the plutonium in the sample. The Pu240 effective mass is derived from the spontaneous fission rate. The total plutonium mass may be determined if the plutonium isotopic ratios are known or can be measured, usually by gamma-ray means.

The triple neutron correlation technique using the Time Correlation Analyser (TCA) allows one to determine the frequency distribution of neutrons in the pulse train arriving from the thermal neutron detector. Three quantities may be derived from experimental neutron frequency distribution data using the point source interpretation model of Hage (Ha85, Ha86, Ci86, Ha92). These three quantities are the effective numbers of singlets R_1, the correlated doubles or pairs R_2 and the correlated triples R_3. Note that R_1 is the Totals count rate and R_2 is the Reals rate familiar in coincidence counting. These measured quantities are related to physically relevant quantities by the well known point source model equations, which are reproduced elsewhere, [Ma93].

The TCA is the basis of data acquisition for the Drum Monitor. The variations of measurement method which are available using the TCA and the point source interpretation model are summarised in Table 1.

Table 1: Measurement and Analysis Methods.

1. Total neutron counting (1 equation in R_1) This is the most sensitive measurement method with the lowest detection limits.

2. Pair Correlation (2 equations in R_1 and R_2)

2.1 **Routine FE**
Known α ratio and multiplication M
Unknown F_S, ε

2.2 **Routine FM**
Known α ratio and detection probability ε
Unknown F_S, M

3. Triple Correlation (3 equations in R_1, R_2 and R_3)
Absolute ^{240}Pu mass determination

3.1 **Routine FEM**
Known α ratio
Unknown F_S, ε, M

3.2 **Routine FESA**
Known Multiplication (M = 1)
Unknown F_S, ε, S_α

3.3 **Routine SAFM**
Known Detection probability ε
Unknown S_α, F_S, M

4. Pair Correlation or coincidence counting using a calibration function.

The prototype and ANTECH commercial variants of the Model 2200 neutron Multiplicity Drum Monitor are 4 π thermal neutron detector systems with 64 He-3 tubes arranged in a decagon. With the internal cadmium liner fitted the maximum detection probability in about 15.5% (JRC prototype) ranging to 20% for the ANTECH Model 2201. The internal cadmium liner may be removed easily for higher efficiency Totals counting.

All of the systems have an external polyethylene shield which is 210 mm thick. Typical neutron decay time for the JRC prototype drum monitor is 70 µsec (67 µsec for the Model 2201). With the internal Cd liner fitted, the detector exhibits a single exponential decay for a period of over 300µ sec.

DRUM MONITOR DESIGN AND DEVELOPMENT

Although the prototype design employed an overhead crane for drum loading, user requirements have dictated the use of a rail system to move drums into and out of the measurement chamber of the Model 2201 Drum Monitors. Drums are placed on the rail system using a manual drum lifter. Other variants of the drum loading system are also possible.

The He-3 detector tubes are organized into 16 detector modules. Each module consists of 4 He-3 tubes (at 4 atmospheres) and a sealed low profile preamplifier-discriminator/high voltage junction box, employing the Amptek model A-111 charge sensitive amplifier. Each of the 16 preamplifier-discriminator outputs are connected to a derandomizing buffer-mixer-counter circuit implemented using VLSI technology. The mixer circuit improves the counting statistics for pair and triple correlation by eliminating coincident count losses. The Mixer circuit has independent counters on each input channel that provide a useful diagnostic feature. The individual mixer channel count totals for each module are monitored by the analysis software. The data will be used in the implementation of the analytic dead time correction which is to be included in the Drum Monitor systems in the near future. The mixer data is fully integrated with the neutron frequency data obtained by the TCA and the system provides a statistical analysis of the count rate seen by each detector module.

Statistical filtering has been implemented to reduce the effect of cosmic ray induced neutron bursts. A variety of filtering criterion are being investigated and a maximum neutron distribution frequency filter is in use at present. The mixer totals counters are also used to provide data for a Fourier analysis of the detector module count rate. This data is used as a means of anomaly detection, for example a significantly non-uniform drum loading of fissile material.

MEASUREMENTS

The results of measurements of the axial variation of the maximum detection probability using a small Cf-252 source and a small Pu sample are plotted in Fig. 1, with no matrix present in the measurement chamber. It can be seen that the detection probability varies as one would expect with the maximum values at the bottom of the chamber for central measurements. The efficiency for the Cf-252 is lower than that for Pu as expected for some of the data. The inconsistency is probably due to poor counting statistics.

Measurements have been made using a well characterized plutonium sample with a known Pu 240 effective mass at different positions on the central axis of the drum with no matrix present. The measurements were repeated using the same Pu sample and measurement positions, however, the matrix was a concrete drum with a central re-entrant tube. Both measurement sets produced acceptable results for the Pu240 effective mass, however, the central measurements in the concrete drum matrix produced significantly larger errors [Ma95]. In Fig. 2 the variations of the measured detection efficiency for the two measurements are plotted as a function of vertical position along the measurement chamber central axis. The effect of the concrete matrix in depressing the detection efficiency is apparent.

Representative data is presented in Table 2 to illustrate absolute measurements using the Drum Monitor for both well characterized Pu and Cf-252 samples. The declared values of the samples are indicated, and the errors have been included. For these example

Fig 1: Drum Monitor Detection Efficiency Cf / Pu

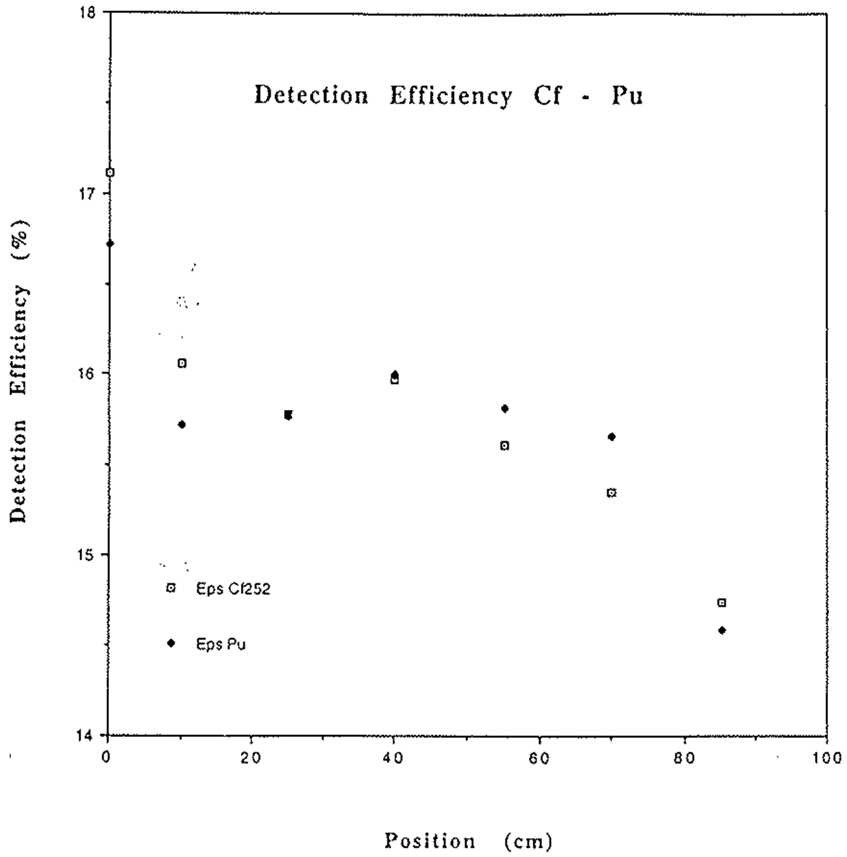

Fig 2: Drum Monitor Detection Efficiency Pu - Empty/Concrete

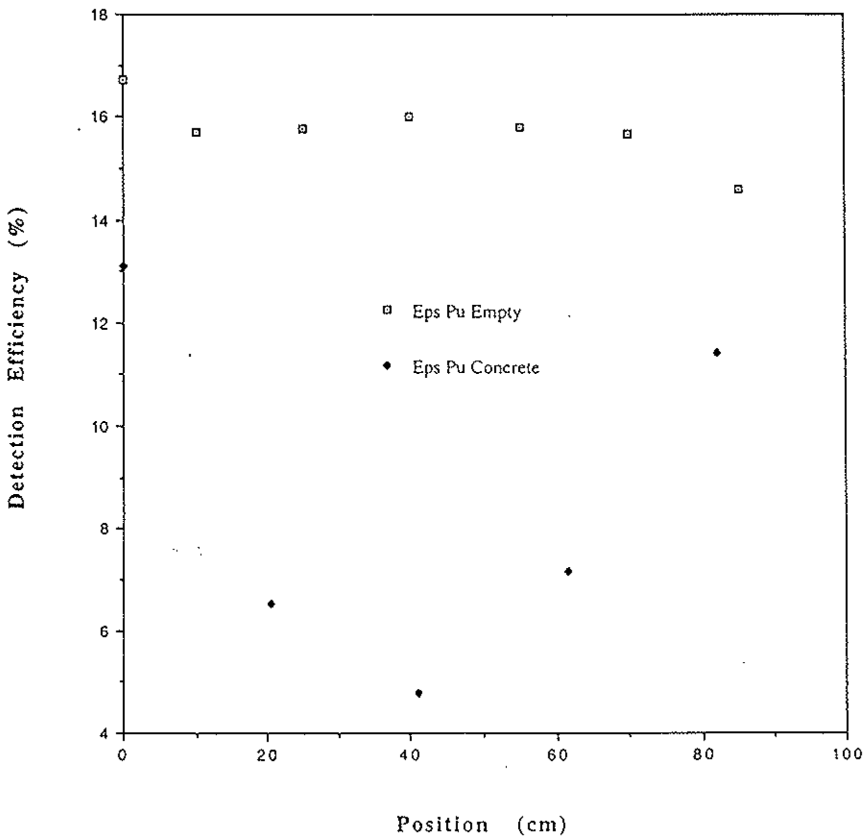

Table 2: Drum Monitor Pu/Cf Measurements

Sample	Count Time (sec)	Count Rate (cps)	Trigger Method	Analysis Method	Fission Rate	MPu240 (g)	M	Alpha Ratio	Epsilon (%)	Diff MPu240	Err MPu240 (%)
Pu22						**2.738**	**1.055**	**0.640**	**15.50**		
	8000	749.6	Combined	FEM	1311	2.737	1.057		15.20	0.001	0.04
				FESA	1286	2.648		0.652	15.48	0.090	3.29
				SAFM	1283	2.679	1.055	0.653		0.059	2.15
			Periodic	FEM	1289	2.690	1.054			0.048	1.75
				FESA	1305	2.723		0.632		0.015	0.55
				SAFM	1298	2.708	1.054	0.635		0.030	1.10
Cf1					**1565**	**-**	**-**	**-**	**13.50**	**Diff (Fs)**	**Err %**
	3600	898.5	Combined	FEM	1557				15.32	8.000	0.51
				FESA							
				SAFM							
			Periodic	FEM	1551				15.37	14.000	0.89
				FESA							
				SAFM							

measurements no matrix was present. Data is included for both the Combined and Periodic Trigger methods as employed in the TCA. For the plutonium measurement, the data has been analysed using all three analysis methods. As expected, good agreement has been achieved for this simple matrix.

No calibration is necessary in these cases as the 'known' parameter is available. In normal operation of the Drum Monitor absolute measurements are made using FESA analysis as it is generally appropriate to assume M = 1 for the waste stream.

CONCLUSIONS

The multiplicity drum monitor has advantages over conventional neutron waste counters. For samples where either the multiplication M or the alpha ratio are known or can be detemined the use of the instrument constitutes an absolute measurement method. Where the known parameter must be estimated, the sensitivity of the Pu mass result to variations of the parameter can be assessed and the potential errors estimated.

Measurements of background counting rates have demonstrated that, provided adequate counting statistics can be obtained, accurate measurements of very small Pu samples (<100 mg) may be made with the multiplicity drum monitor. Further work is currently being undertaken to improve the measurement technique. The module and junction box design is being revised to increase the detection efficiency and enhance performance. Improvements are also being made in the assay software to improve the diagnostic information available to the operator.

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