SPENT FUEL AND RESIDUE MEASUREMENT INSTRUMENTATION AT THE SELLAFIELD NUCLEAR FUEL REPROCESSING FACILITY

Andrew S Chesterman
& Peter A Clark
BNFL Instruments Ltd., Pelham House, Calderbridge, Seascale,
Cumbria CA20 1PG, United Kingdom. Tel: +44(0) 19467 75654 Fax: +44(0) 19467 76579

ABSTRACT

The Sellafield reprocessing plant receives and reprocesses several thousand tonnes of spent light water reactor (LWR), advanced gas cooled reactor (AGR) and natural uranium magnesium alloy clad (Magnox) fuels each year. The safety and cost effectiveness of these operations has been supported by the development and installation, at key points in the process, of a range of special purpose radiometric instrumentation. Systems in routine operational use verify the cooling time, burn-up and initial and final U-235 equivalent enrichment of fuel assemblies in the storage and handling ponds. Other systems determine the radionuclide inventories of fuel residues in intermediate level waste arising from plant operations. The measurement techniques employed include high resolution gamma spectrometry, passive neutron counting and neutron interrogation by the use of a Cf-252 source and deuterium-tritium (D-T) pulsed neutron generators. Details of the instruments including mechanical installation arrangements and measurement data are presented in the paper along with a discussion of possible future uses of similar instruments for burn-up credit associated with fuel and residue storage, transportation and disposal.

INTRODUCTION

The growth over the last two decades of spent fuel reprocessing at Sellafield has demanded a parallel growth in the development and use of specialised radiometric instrumentation. The impetus for this growth has come from the requirements of plant operators, the Safeguards Authorities (IAEA and Euratom) and the UK nuclear waste agency (NIREX). Their respective requirements are to have on-line information of plant processes to ensure safe and efficient operation in accordance with the nuclear site licence, to track the movement through the plant and measure the quantities of special nuclear materials (SNM), principally fissile uranium and plutonium, and to measure radionuclides within process residues and wastes which are destined for disposal in an underground repository. In addition instrumentation with improved accuracy and sensitivity is also required to support decommissioning of redundant plant and for spent fuel storage and transportation.

Predominantly the development, installation, commissioning and operational support of radiometric instruments at Sellafield have been carried out by an "in-house" team of about 90 physicists and engineers. This team has now formed the core of the new company, BNFL Instruments Ltd.

From a total of more than a hundred instrument systems developed and installed in operating plant by the team, this paper describes four:

- The Short Cooled Fuel Monitor
- The Swarf Inventory Monitor
- The Feed Pond Fuel Monitors
- The Hulls Monitor

The selected instruments are used for spent fuel and intermediate level waste measurements in the mechanical head end of the two reprocessing facilities at Sellafield; the Fuel Handling Plant (FHP) and the Thermal Oxide Reprocessing Plant (THORP). The FHP is a decanning facility for natural uranium magnesium alloy clad (Magnox) fuels. THORP is a reprocessing facility for oxide fuels, primarily from LWRs and AGRs, and its functions include mechanical handling, shearing and chemical separation with a maximum throughput of 1200 tonnes uranium per year.

The design process for the four instruments were similar in that they all adhered to a strict quality assurance procedure which is formalised by the joint production, between the customer and design team, of a Measurement Specification. This document identifies the performance requirements, the installation constraints and plant interface details. The instrument specification, based upon previous experience or on specific development work, is documented by the design team in the System Specification and is issued to the customer for approval prior to component ordering, manufacture and assembly of the final instrument. Finally a commissioning program is drawn up and implemented by the design team in conjunction with the customer. This joint team approach is an important part of the instrument's transition to operational status.

The selected instruments are all controlled by in-built computer systems to facilitate measurement sequencing, data processing, automatic standardisation routines, fault diagnosis and communications between

the instrument, plant PLCs and operators. The radiometric measurement techniques employed are one or more of; high resolution gamma spectrometry (HRGS), passive neutron measurements and active neutron measurements.

THE FUEL HANDLING PLANT COOLING TIME MONITOR

Background - The role of the FHP cooling time monitor is to confirm that Magnox fuel elements fed to the decanning line are greater than 150 days cooled. This ensures that the ^{131}I ($t_{1/2}$ 8.04 days) has decayed to insignificant levels. Historically a relatively simple instrument based on low resolution gamma spectrometry and the detection of ^{140}La ($t_{1/2}$ 1.68 days daughter of ^{140}Ba $t_{1/2}$ 12.7 days) was devised for fuel cooling time measurements. For the FHP a more sophisticated cooling time monitor was developed to provide quantitative information on both cooling time and burn-up by using HRGS to measure a number of fission product ratios and absolute ^{137}Cs activity.

The mechanical arrangement of the FHP cooling time monitor, figure 1, shows the position of the roof mounted collimator plug which allows the coaxial intrinsic Ge detector to view the individual Magnox fuel rods when they are rolled into the v-shaped holder in the operations cave 4m below. After successful measurement the rods are pushed by a hydraulic ram through a slitter head which peels the Magnox cladding from the spent fuel bar. The removed cladding (swarf) falls onto a collecting tray for further measurements by the Swarf Inventory Monitor.

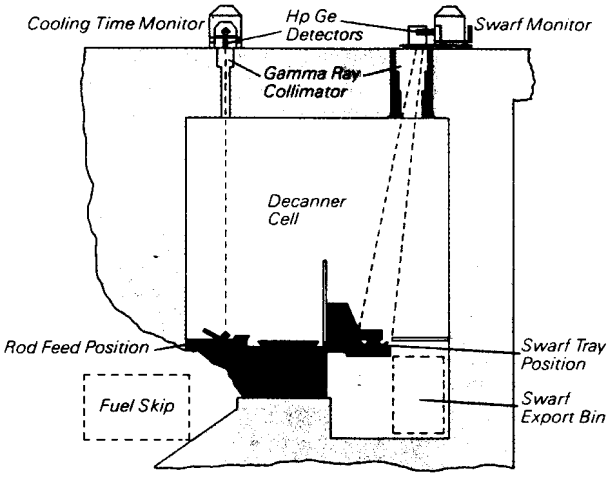

Figure 1 Mechanical Arrangement of the FHP Cooling Time Monitor and Swarf Inventory Monitor.

Radiometric Method - The cooling time is determined using the activity ratios $^{140}La/^{95}Zr$ and $^{95}Zr/^{144}Ce$. The general criteria for selecting activity ratios for cooling time determination are that they should ideally be independent of irradiation history, vary as strongly as possible with cooling time and be easily measurable. It follows that the chosen nuclides should be in a state of equilibrium when the spent fuel is released from the reactor, the two nuclides in the ratio should have significantly different half lives and their gamma emission photopeak energies must be measurable and not interfered with by other photopeaks in the fuel gamma spectrum. For normal dwell times the nuclides ^{140}La ($t_{1/2}$ 12.7 days), ^{95}Zr ($t_{1/2}$ 64 days) and ^{144}Ce ($t_{1/2}$ 285 days) are in equilibrium and therefore are independent of irradiation. Also the half lives are sufficiently different to give 1σ error bars, based on 140 days cooled fuel and a 60 second measurement time, of about ± 3 days from $^{140}La/^{95}Zr$ and ± 8 days from $^{95}Zr/^{144}Ce$. Due to nuclide decay the ratios $^{140}La/^{95}Zr$ and $^{95}Zr/^{144}Ce$ are useful only for fuels cooled to about 160 and 500 days respectively.

The irradiation is determined by using either the activity ratio $^{106}Ru \times {}^{137}Cs/({}^{134}Cs)^2$, which has an effective half life of 22 years, or the absolute activity of ^{137}Cs. These both fulfil the criteria for irradiation parameters namely that they are only weakly dependent on cooling time and reactor rating.

THE FUEL HANDLING PLANT SWARF INVENTORY MONITOR

Background - The Swarf Inventory Monitor (SIM) uses HRGS and views the Magnox swarf on a sorting tray through a roof collimator, figure 1. Measurement of spent uranium fuel mass and the radionuclide inventory associated with the swarf is made immediately prior to the swarf being pushed into a swarf bin. If a fuel mass greater than a reference limit is detected then a "high fuel" alarm is given to the operators to prompt the removal of any identifiable pieces of fuel. Any retrieved pieces are re-routed to join the decanned fuel stream for chemical separation, thereby minimising fuel losses with the waste swarf. Following a high fuel alarm the monitor will automatically request a repeat measurement to determine the remaining nuclide inventory on the tray. When a full swarf bin is exported to the adjacent waste encapsulation plant the monitor integrates the total inventory assigned to the bin. The amount of uranium carried with the waste is used as an entry into the SNM account.

Radiometric Method - The uranium fuel mass and radionuclide inventory are calculated from the measured activities of the radionuclides listed in table 1. These along with other fission product radionuclides entrained in fuel fragments and ^{60}Co activation in steel components such as nimonic springs give rise to the

observed gamma spectra. Having corrected the gamma spectra for background activity determined from an empty tray measurement following each swarf measurement, the net photopeaks, within the energy range 500 - 1600keV, are determined.

Table 1 Radionuclides measured by the SIM

Nuclide	Half-life	Energy of principal gamma rays (keV)	Branching. ratio (%)
^{95}Zr	64.0d	724.2	44.1
		756.7	54.5
^{95}Nb	35.0d	765.8	99.8
^{106}Ru(Rh)	1.02y	511.9	20.7
		621.9	9.8
^{134}Cs	2.062y	569.3	15.4
		604.7	97.6
		795.9	85.4
		802.0	8.73
		1365.2	3.04
^{137}Cs	30y	661.7	85.2
^{144}Ce(Pr)	285d	696.5	1.34
^{154}Eu	8.8y	723.4	19.7
		1004.8	17.9
		1274.5	35.5

A relationship between irradiation and ^{137}Cs specific activity (Bq/g(U)) derived from the nuclide inventory code, FISPIN[1], is used with the measured ^{137}Cs and irradiation determined from the ^{106}Ru x ^{137}Cs/(^{134}Cs)^2 activity ratio to give fuel mass.

The cooling time, which is needed to make a small correction to the irradiation and in the inferential determination of the non measurable fission product radionuclides, is calculated from the two ratios ^{95}Zr/^{144}Ce and ^{134}Cs/^{137}Cs. The activities of 29 non measurable radionuclides are inferred by the use of look-up curves which correlate their activity with that of ^{137}Cs and the measured irradiation and cooling time.

Other features incorporated in the radiometric method are an energy dependent relative gamma detection efficiency correction and a self attenuation correction to correct the ^{137}Cs photopeak detection efficiency for the effect of different sizes of fuel debris. This latter routine uses a "Newton-Raphson" iterative technique which gives a representation of the distribution of fuel sizes present on the tray by a mix of two particles, one of zero thickness, i.e. fines offering no attenuation to the emitted gammas, and the other of a finite thickness up to the approximate diameter of a fuel bar (30mm).

The results of a performance assessment have shown that for over a million rods measured to date the accuracy of fuel mass determination as assigned to the exported swarf bins is in the range of ± 8%.

THE THORP FEED POND FUEL MONITORS

Background - The primary role of the Feed Pond Fuel Monitors (FPFM) is to ensure that all fuel entering the Head End of THORP for reprocessing conforms to the plant flow sheet acceptance criteria. For the initial THORP base load fuels the acceptance criteria are; LWR fuel irradiation ≤ 40 GWd/Te(U) and cooling time ≥ 5 years, AGR fuel irradiation ≤ 25 GWd/Te(U) and cooling time ≥ 3.8 years and for both LWR and AGR fuels pre-irradiation initial enrichment ≤ 4% ^{235}U. The requirements of the monitor are therefore to measure these parameters, without reference to operator declared fuel data other than fuel type, and to give a "Go" or "No Go" signal dependent on whether they are within the acceptance envelope. In addition the monitor is to measure the fuel final enrichment, as % ^{235}U equivalent

To meet a throughput of 7 Te(U) per day two Fuel Removal Machines (FRMs) and two associated monitoring stations were installed, figure 2. Each FPFM employs a 15% efficiency high purity germanium gamma detector, and five fission chamber neutron detectors that are split into two modules arranged at 90^o to each other. A neutron source transfer system, controlled by the FPFM, moves a ^{252}Cf source to an exposed and shielded position to allow active and passive neutron measurements. The exposed source, which is located in a third module opposite the three detector module, is also used during standardisation to check the counting efficiency of the neutron detectors. The position of the detector and source modules are adjustable to accommodate a range of fuel sizes. The gamma detector is standardised with the use of a mixed ^{137}Cs and ^{134}Cs source moved by an electromechanical actuator between a shielded position in the detector housing to an exposed position adjacent to the detector crystal. The gamma detector front end electronics comprise an EHT power supply, gated integrator amplifier and analogue to digital converter (ADC). The digital signal from the ADC is fed to a loss free counting module and a MicroVAX based multichannel analyser via an Ethernet link. The analogue signals from the neutron detector amplifier/discriminators are converted to digital TTL pulses and fed to separate counter and timer cards in a VME computer system. Preliminary data processing for the neutron counting system is performed by the VME computer system and all other functions including plant communications are carried out by the MicroVAX.

Measurement Procedure - While a fuel item is being removed from its storage container the selected monitor is standardised. Measurements are then made at each of up to four measurement heights during the

rotation of the fuel in the monitor station. The local values of cooling time, irradiation, initial and final enrichments are calculated at each measurement height together with the weighted average for all measurements on the assembly. The weighted average values are used to generate a Go or No Go signal. If this is Go the assembly is delivered to an elevator for transit to the shear cave. If the signal is No Go the assembly is returned to its storage container for further investigation.

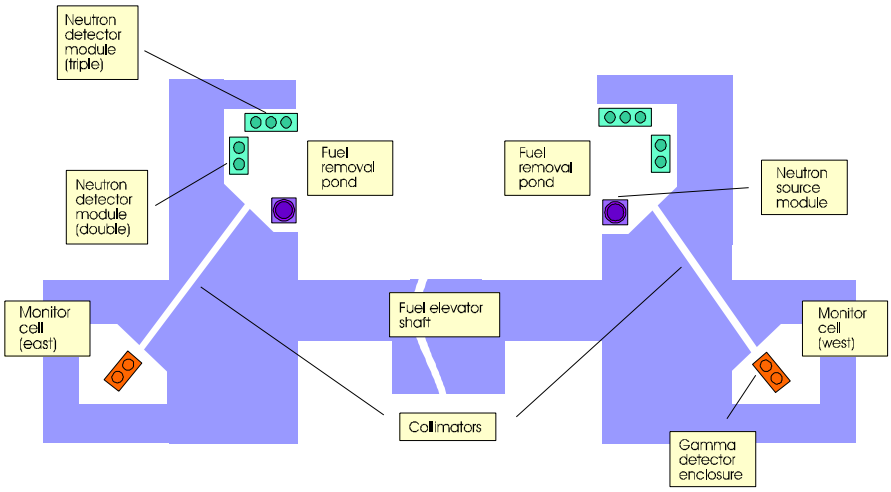

Figure 2 Schematic Plan View of the THORP Feed Pond Fuel Monitors

Radiometric Method - A combination of three techniques are used comprising HRGS, passive neutron emission measurement, and the measurement of the neutron emission from fissions induced by an external ^{252}Cf neutron source. In the gamma spectrometry procedure the net photopeak areas of the selected nuclides, ^{137}Cs, ^{134}Cs, ^{106}Ru and ^{154}Eu, are corrected for background activity and normalised for relative detection efficiency using the multiple photopeaks of ^{134}Cs.

Cooling Time - Two independent values of cooling time are determined by HRGS using the activity ratios, ^{134}Cs/^{154}Eu and ^{106}Ru/^{137}Cs. The final cooling time for a particular measurement position is then derived from a weighted mean of the two values. The weights being inversely proportional to their associated errors.

Irradiation - Three separate values of irradiation are determined at each measurement position; two use HRGS and the third uses the passive neutron measurements. In the HRGS determination the absolute ^{137}Cs count rate and the activity ratio (^{106}Ru x ^{137}Cs)/(^{134}Cs)^2 are used. The irradiation has a linear relationship with ^{137}Cs and a log relationship of the following form with the activity ratio; Ln(I) = m . Ln(R_0) + c, where I = the irradiation, R_0 = the measured activity ratio corrected to zero cooling and m and c are calibration constants. The third method for irradiation determination makes use of the following relationship between passive neutron emission, principally from ^{244}Cm, and irradiation; Ln($N_{p(0)}$) = a . Ln(I) + b, where $N_{p(0)}$ = zero cooled background corrected passive neutron count rate and a and b are enrichment dependent calibration terms appropriate to each fuel type. The final irradiation value is calculated by taking a weighted mean of the three independently measured values.

Final Enrichment - The final enrichment, expressed as %^{235}U equivalent, is determined by a combination of the measured irradiation and a neutron multiplication parameter from the active neutron measurements using the external ^{252}Cf neutron interrogation source. The relationship linking final enrichment F and the multiplication M is; M = r . F + z, where r is an irradiation dependent calibration term for each fuel type and z is a fuel dependent zero fissile term.

Initial Enrichment - The initial enrichment, En, is calculated from a combination of the final enrichment and measured irradiation via FISPIN[(1)] calibration data. The relationship is of the form; En = y(I) . F, where y(I) is an irradiation dependent, calibration term.

Fuel Measurements - Examples of calibration measurements for BWR fuel are shown in figures 3, 4 and 5. Figure 3 shows a BWR fuel cooling calibration based on the measurement of 24 assemblies. The standard deviation of the fit is 83 days.

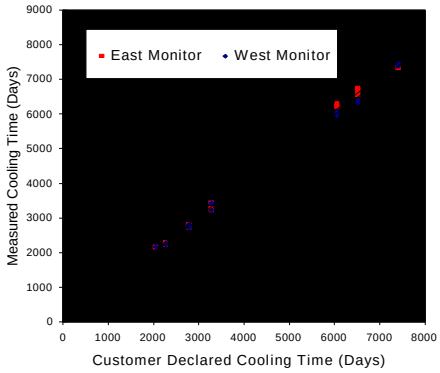

Figure 3 Measured and customer declared values of cooling time for BWR fuel calibration data.

Figure 4 shows the irradiation determined by a combination of the ^{137}Cs and activity ratio methods. The fit has a standard deviation of 1.0 and 0.8 GWd/Te for

the west and east monitoring stations respectively

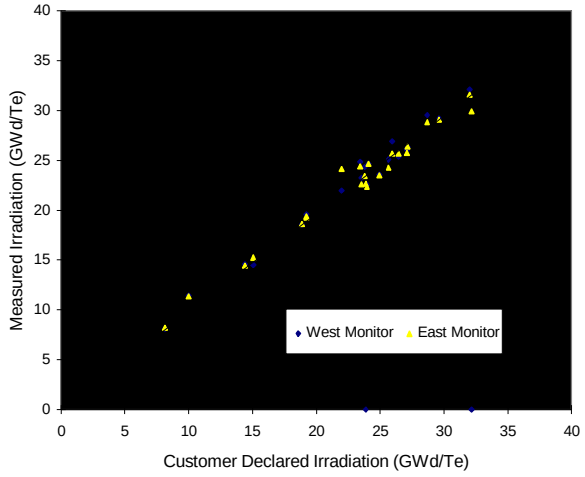

Figure 4 Measured and customer declared values of irradiation for BWR fuel calibration data.

.

Figure 5 shows the initial enrichment, determined by the west monitor station, this has a standard deviation of 0.12% ^{235}U.

Further irradiation results from 80 BWR fuel assemblies measured during the first THORP campaign of BWR fuel are shown in figure 6. These are based on a combination of the absolute ^{137}Cs and activity ratio method of irradiation determination and have a standard deviation of 0.48 GWd/Te.

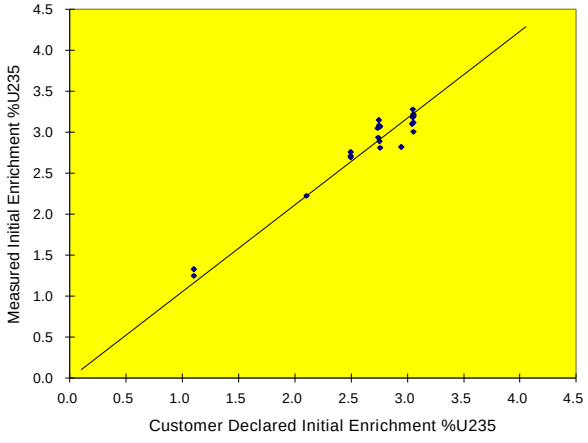

Figure 5 Measured and customer declared values of initial enrichment for BWR fuel calibration data

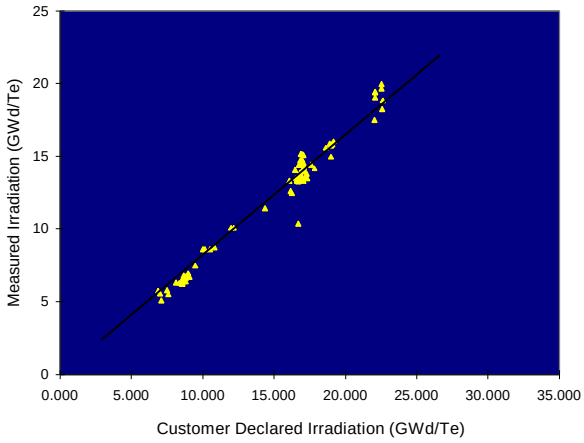

Figure 6 Measured and customer declared values of irradiation for 80 assemblies from the first THORP campaign of BWR fuel.

THE THORP HULLS MONITOR

Background - After dissolution of sheared fuel in THORP the resulting pieces of empty fuel cladding (hulls) are measured for process and criticality control purposes prior to encapsulation for disposal. A schematic layout of the hulls dissolver basket and the radiometric equipment is shown in figure 7. The measurement takes place on the basket, which has a diameter 700mm and maximum fill height of 2300mm, as it is moved vertically in a spiral manner through the measurement collar. The Hulls Monitor measurements are required; (i) to ensure criticality control during the movement, encapsulation and storage of the hulls, (ii) to determine the remaining quantities of spent fuel including the amounts of fissile material (uranium-235, total uranium and fissile and total plutonium) for process control, accountancy and quality assurance, and (iii) to measure a radionuclide inventory of the basket contents which are eventually tipped into a 500 litre drum for cement encapsulation prior to consignment to a repository.

Radiometric Method - The Hulls Monitor uses three radiometric techniques; (i) Differential Die Away (DDA) for the determination of the ^{235}U equivalent residual fissile content, (ii) passive neutron counting for residual fuel content, and (iii) HRGS to give an independent determination of residual fuel and radionuclide inventory of the gamma emitting fission and activation products.

The DDA neutron interrogation technique is becoming increasingly important for a variety of fissile material measurement applications, and was considered the most appropriate technique for the Hulls Monitor

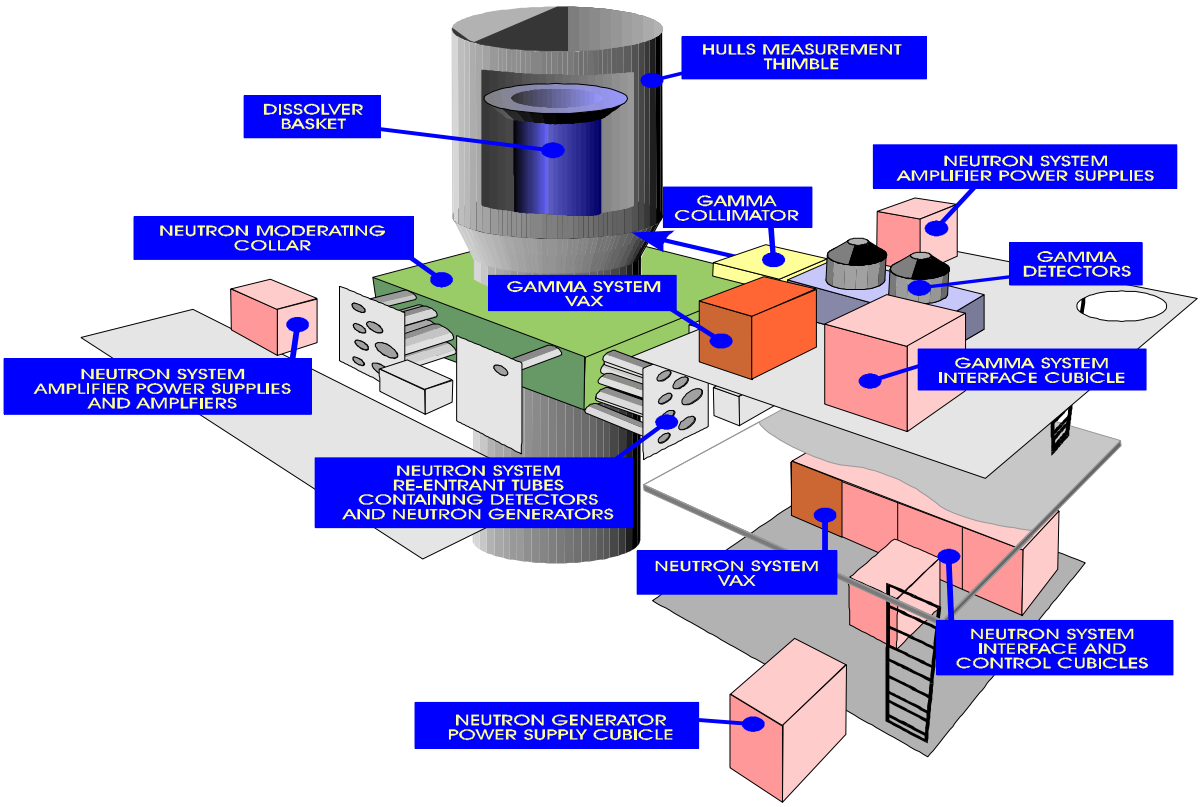

Figure 7 Schematic Arrangement of the Hulls Monitor.

because the passive radiation emissions from the leached hulls do not provide a unique measure of fuel content. In the Hulls Monitor 14MeV neutron pulses of 90µS width at 15Hz, generated by a deuterium-tritium (D-T) neutron generator, are injected into the cavity around the measurement collar. After the initial burst of neutrons

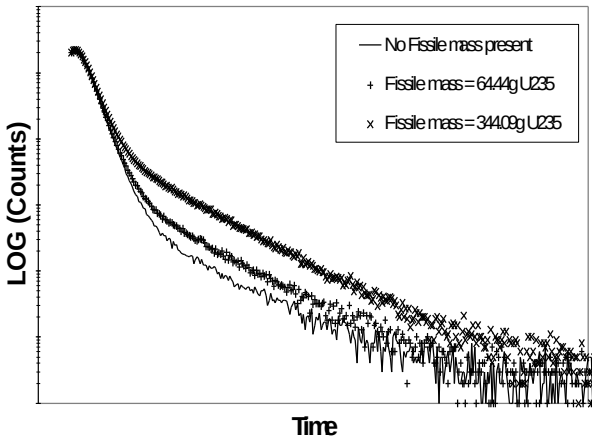

Figure 8 Decay of fast neutron flux in Hulls Monitor DDA measurement cavity.

from each generator pulse the fast neutron flux within the measurement cavity quickly dies away as the neutrons are thermalised, absorbed or escape from the cavity. The rate of neutron flux decay is slowed, however, by the presence of fissile material in the cavity due to neutron production from induced fissions. This allows a measure of fissile mass by counting the integrated fast neutron flux in the die away period. Figure 8 shows examples of fast neutron flux decay.

To determine the residual fuel content passive neutron counting, primarily of the ^{244}Cm spontaneous fission neutrons, is used with the DDA measured fissile content and the initial enrichment and cooling time derived from the Feed Pond Fuel Monitor measurements. The technique uses the ratio of the cooling corrected passive neutron emission and fissile content to give a parameter related to the mean irradiation of the measured material. This together with a knowledge of initial enrichment allows the fuel content per gram of fissile to be found and hence the total fuel mass. A plot of measured fissile mass against known mass values from fissile standards is shown in figure 9. The measurement errors, 30% at 3σ, are dominated by systematic errors due to fuel positioning and matrix variations. The detection limit for fissile mass is typically 6g 235U equivalent fissile in a 600kg batch of leached BWR hulls.

The HRGS measurements provide an independent fuel content measurement using ^{134}Cs, ^{137}Cs and ^{154}Eu and therefore give a diverse indication of gross dissolver maloperation. In addition the gamma spectra

give a direct indication of the amounts of "measurable" radionuclides.

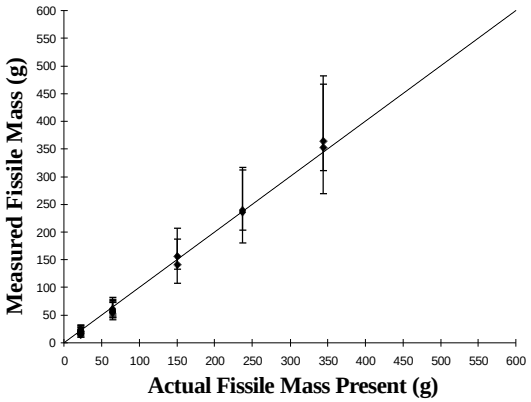

Figure 9 Hulls Monitor DDA calibration measurement check

A plot of measured against known radionuclide activities from destructive analysis, is shown in figure 10. Again as for the fissile mass measurements the errors are dominated by systematic errors from positioning and matrix variations. Other non-measurable radionuclides are inferred from the DDA fissile mass combined with the passive neutron count, the cooling time, irradiation and fuel inventory code data to give a total radionuclide inventory

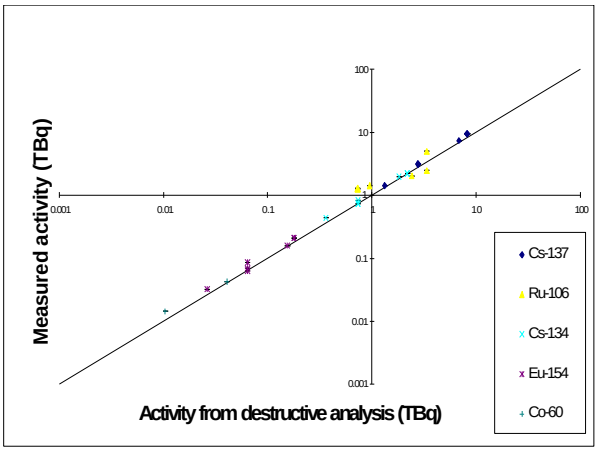

Figure 10. Gamma system results measuring spent fuel standards.

DISCUSSION AND FUTURE APPLICATIONS

The experience gained by the development and industrial use of a wide range of radiometric instruments is supporting a development program aimed at meeting actual and anticipated instrumentation requirements. Examples now under development include capabilities for the measurement of:

- High burnup and mixed oxide fuels for THORP reprocessing.
- Wastes from decommissioning and retrieval operations.
- Spent fuel for burn-up credit requirements.

The need for burn-up credit measurements is becoming increasingly important as storage space at many reactor sites is filled. The requirement to increase the effective capacity of transport containers is also economically important. This is emphasised by the fact that there are currently a number of spent fuel transport casks being designed with an intention to utilise burn-up credit in their loading arrangements[(2), (3), (4)]

As the instrumentation requirements for burn-up credit are closely related to existing devices such as the Feed Pond Fuel Monitors, burn-up credit is regarded as a priority area for product development. The primary objective is to make available during 1996 a commercial burn-up credit monitor (BCM). This device will have various configurations, one of which is illustrated in figure 11.

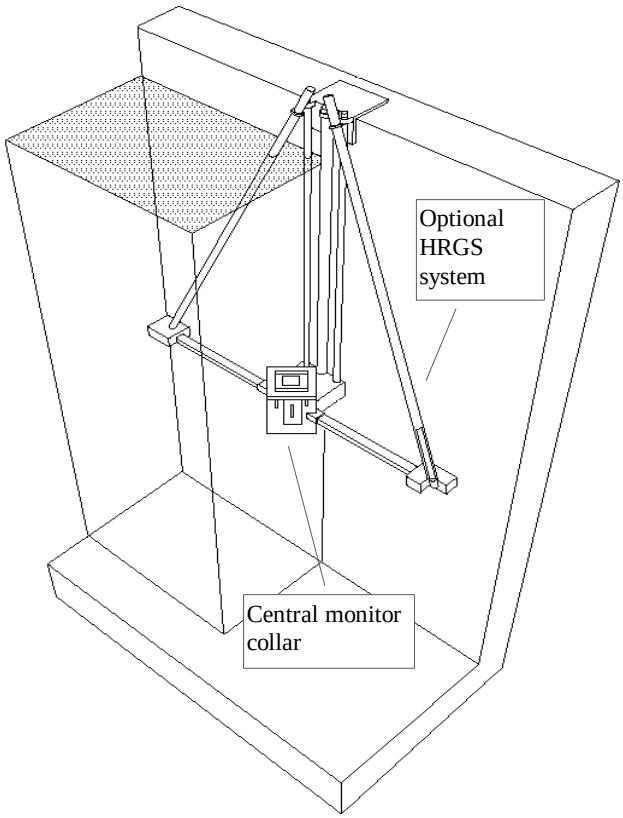

Figure 11 Burn-up credit monitor configuration.

The basic components will comprise; a central monitor collar which will contain four fission chamber neutron detectors, a ^{252}Cf interrogation source and two ion chambers. The neutron detectors will be placed symmetrically about the fuel monitoring vertical axis. This central collar will therefore offer passive neutron measurements, active neutron measurements and total gamma counting. Options will include the ability to measure at one or more positions along the length of the fuel assembly or a continuous measurement as the assembly is lowered or raised through the central collar. If the continuous measurement option is selected the ion chambers would trigger the start and stop of the measurements as the fuel passes through the collar. A further option will be a HRGS capability with the use of two Ge detectors loaded via the diagonal re-entrant tubes into shielded enclosures at the bottom of the tubes. These will view the fuel through two horizontal gamma collimators attached to the central collar.

The main structural components will be fabricated using stainless steel with a high density polyethylene insert within the central collar to aid fuel location and minimise frictional contact. The construction will be modular to allow easy transportation and on-site assembly. An objective of the design is to produce a device which can be installed into an existing plant or storage pond with minimum disruption.

Techniques for the interpretation of the measurement data are being optimised. These include; (i) the use of the passive neutron measurements to give a confirmatory check of the operator declared fuel data, (ii) built in compensation for variable measurement conditions, for example boron doped pond water, (iii) relating the multiplication parameter from the active neutron measurement to the cask k-effective value, and (iv) extraction of burnup, cooling time and enrichment from the gamma spectrometry measurements.

A wide range of spent fuels are available at Sellafield for proving and optimising the design of the BCM to provide a low maintenance and robust device for industrial applications where high reliability is an essential attribute.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the contribution made by the following BNFL Instruments Ltd personnel who have significantly contributed to the development and commissioning of the above instruments; A D Messado, I J Casson, J Rackham and N H Merrill.

REFERENCES

(1) FISPIN - A computer code for nuclide inventory calculations, R F Burstall, United Kingdom Atomic Energy Authority, ND-/R/328(R), October 1979.

(2) Burnup Credit Experiences with the GA-4 Cask, Jack K Boshoven, Nuclear Technology Vol.110, p.33, April 1995.

(3) Feasibility Assessment of Burnup Credit in the Criticality Analysis of Shipping Casks with BWR Spent Fuel, Bryan L Broadhead, Nuclear Technology Vol.110, p.1, April 1995.

(4) Evaluation of Burnup Credit for Fuel Storage Analysis - Experience in Spain, Jose M Conde and Manuel Recio, Nuclear Technology Vol.110, p.22, April 1995.