MULTIPURPOSE IN-LINE SPECIAL INSTRUMENTATION IN SPENT FUEL RECYCLE PLANTS

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ABSTRACT

Safe and optimum operations of spent fuel recycle plants rely on the availability of real time measurement systems at key in-line points in the process. More than thirty types of such special instrument systems have been developed and commissioned on the THORP reprocessing plant at Sellafield. These systems are reviewed together with associated information on measurement purpose, measurement technique and plant performance. A number of these measurement systems also support Materials Accountancy and Safeguards arrangements on the plant.

A more detailed overview of two such instrument systems within the Head End and Product Finishing Stages of THORP is provided. The first of these is the Hulls Monitor, based on active and passive neutron measurements, as well as high resolution gamma spectrometry, of the basket of leached fuel cladding. This provides vital data for criticality assurance, nuclear material accountancy and inventory determination for ultimate disposal of the cladding waste. The second system is a Plutonium Inventory Monitoring System (PIMS) which employs passive neutron counting from a distributed array of neutron detectors within the Pu Finishing Line. This provides a near real time estimate of Pu inventories both during operations and at clean out of the Finishing Line. Both the Hulls Monitor and PIMS technologies are applicable to MOX Fuel recycle. Both systems enhance the control of fissile material in key areas of the recycle process which are of interest to the Safeguards authorities.

INTRODUCTION

The benefits of in-line radiometric instrumentation are well established for the safe and optimised operation of spent fuel recycle plants. The measurement of process material within the plant vessels and containment eliminates many of the systematic errors, time delays and waste arisings associated with sampling and off-line analysis. The real time nature of in-line measurements using computer controlled data acquisition and processing has enabled this type of special instrument to fulfil key control and monitoring needs on successive generations of recycle plants at Sellafield.

Where the required in-line measurement systems are not commercially available, the lifetime costs of designing, installing and operating appropriate special instruments have to be justified in terms of the benefits of their use. The purpose of an instrument may be for process monitoring/control, criticality control, safeguards, materials accountancy or waste stream characterisation. Clearly justification is assisted where the instrument serves more than one purpose. The principles adopted at Sellafield to maximise the benefits of a special instrument system include:

(i) fully integrating the instrument into both the physical plant design and into the operational quality plan/safety case.

(ii) designing the instrument using hardware of known reliability with high integrity software based instrument supervision and self-checking to identify fault conditions.

(iii) investigating the instrument performance such that its operating envelope and accuracy are known and can be confirmed during commissioning.

(iv) using feedback on instrument performance on plant to generate improvements in future applications.

RANGE OF SPECIAL INSTRUMENTS ON THORP

More than 30 types of special instrument systems were developed and commissioned on the THORP reprocessing plant during the 1980s and early 1990s. In many cases designs were built on the operating experience of similar types of instrumentation previously installed on the Magnox reprocessing facilities at Sellafield. Table 1 provides a plant wide overview of the radiometric special instruments on THORP. The instruments are grouped according to plant area and for each system the following are compiled

- parameter(s) measured
- purpose of the measurement
- technique(s) utilised
- achieved performance

Feedback on operational performance of these instruments is regularly reviewed to improve future applications of the technologies as standard commercially available instrument systems.

Two of the THORP special instruments which provide information of interest to the Safeguard authorities are studied in more depth as case examples.

CASE EXAMPLE 1: THE THORP HULLS MONITOR

Background

After dissolution of sheared fuel the resulting pieces of empty fuel cladding (hulls) are measured by the Hulls Monitor prior to sentencing for export for encapsulation and disposal.

Hulls from each dissolver batch are measured in a dissolver basket, which has a diameter 700mm and maximum fill height of 2300mm, as shown in figure 1. The Hulls Monitor measurements are required;

(i) To assure criticality safety of the hulls during subsequent handling in THORP and the Waste Encapsulation Plant.

(ii) To provide process control data on the leach efficiency to permit sentencing of each hulls batch.

(iii) To determine the residual masses of ^{235}U, total uranium and fissile and total plutonium in the hulls for materials accountancy and International Safeguards purposes.

(iv) To provide an activity inventory for compliance with Intermediate Level Waste contractual agreements and repository acceptance criteria.

Radiometric Technique

The Hulls Monitor uses three radiometric techniques; Differential Die Away (DDA), passive neutron measurement, and High Resolution Gamma Spectrometry (HRGS). The DDA measurement determines the residual fissile content of the hulls while HRGS gives the radionuclide inventory of the gamma emitting fission and activation products and provides an independent indication of gross dissolver maloperation.

For the DDA measurement 14MeV neutron pulses of 90µS duration at 15Hz, generated by a deuterium-tritium (D-T) neutron generator are injected into a neutron moderating collar as the dissolver basket is rotated and lowered through the collar. After each pulse of neutrons, the fast neutron flux within the measurement cavity quickly dies away as the neutrons are thermalised, absorbed or escape. Any fissile material in the cavity slows the decay of the fast neutron flux as a result of 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 2 shows examples of fast neutron flux decay in irradiated fuel hulls batches containing 3g and 250g ^{235}U equivalent residual fissile content.

PLANT AREA	MEASUREMENT REQUIRED	PURPOSE OF MEASUREMENT	TECHNIQUES
Receipt and Storage	Verification of spent fuel in multi-element bottles (MEBs)	Process control & Safeguards	Low resolution gamma spectrometry
Feed Pond	Level of MEB flush water in dilution vessel.	Process control	Neutron transmission
	Monitoring of spent fuel assemblies for cooling time, initial enrichment and irradiation	Criticality, process control and International Safeguards	HRGS, passive neutron a active neutron
Reagent make-up	Concentration of gadolinium in acid feed to dissolvers and in shear pack wash water.	Criticality and process control	Neutron transmission
Basket handling cave	Residual fissile, fuel and activity inventory of leached fuel hulls in a dissolver basket.	Criticality and process control, materials accountancy, International Safeguards and characterisation of hulls waste	Neutron interrogation, passive neutron counting and HRGS
Chemical Separation	Pu concentration in various process streams.	Criticality and process control	Passive neutron and XRF
Pu Finishing	Pu mass and distribution throughout finishing line (PIMS).	Process control and safeguards	Passive neutron
	PuO_2 content of product cans.	Safeguards and materials accountancy	Neutron coincidence counting and HRGS
	PuO_2 level in various hoppers and in product can.	Process control	Gamma transmission

Table 1 **Overview of THORP In-line Radiometric Special Instruments**

PLANT AREA	MEASUREMENT REQUIRED	PURPOSE OF MEASUREMENT	TECHNIQUES
Pu Finishing (contd.)	Pu concentration in product from OML evaporator.	Criticality and process control	Passive neutron
	Pu concentration in OML evaporator overheads and steam condensate.	Process control	Low resolution X ray spectrometry
	Plutonium content of waste packages at point of origin.	Criticality and process control	Total neutron counting in mobile system
	Plutonium content of waste packages at central waste facility	Criticality/process control and waste specification	Neutron coincidence counting and HRGS
U Finishing	Enrichment of uranyl nitrate	Criticality and process control	HRGS and gamma transmission
	Enrichment of UO_3 in product drums	Criticality and process control	HRGS
Ventilation and off-gas	Alpha and beta activity on particulates	Process control and monitoring	Scintillation counting
	Kr-85 in dissolver off-gas	Process monitoring and discharge accountancy	LRGS
	Ru-106 in DOG and vessel vent	Process control and monitoring	LRGS
	I-131 in DOG	Process control and monitoring	LRGS

Table 1 (contd.) **Overview of THORP In-line Radiometric Special Instrume**

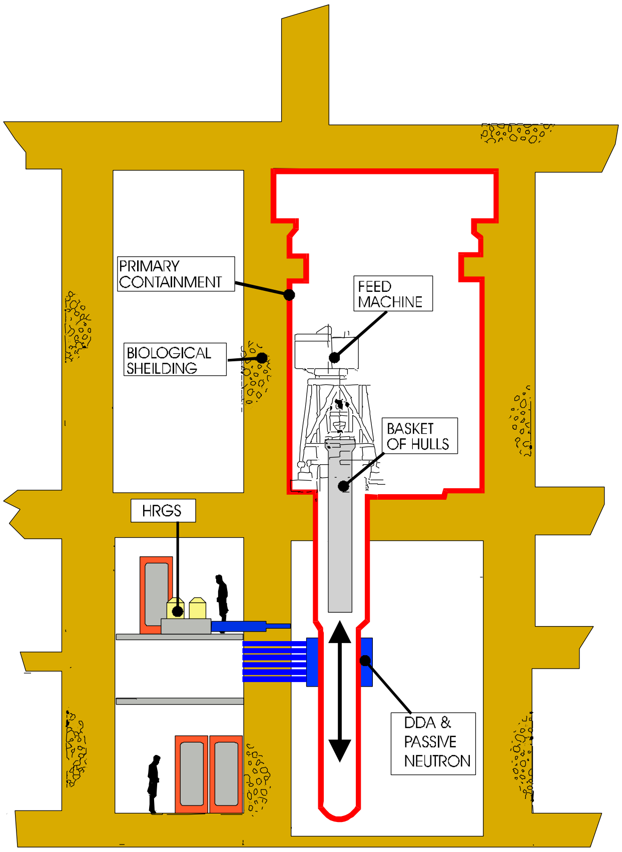

Figure 1 Schematic Arrangement of the Hulls Monitor.

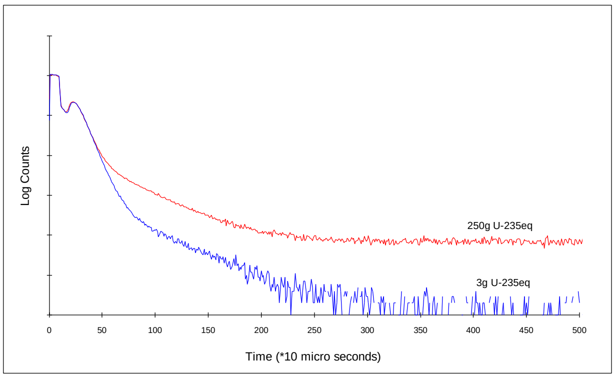

Figure 2 Decay of Fast Neutron Flux in Hulls Monitor DDA Measurement Cavity

The passive neutron emission rate, comprising primarily ^{244}Cm spontaneous fission neutrons, is measured as the dissolver basket is raised through the moderating collar. The passive neutron emission rate is used with the measured fissile mass and the initial enrichment (from the Feed Pond Fuel Monitor measurement before shearing) to determine the residual uranium mass.

The ^{244}Cm activity, total Pu activity, total U activity, total alpha activity, fissile Pu mass, total Pu mass and ^{235}U mass are also determined from the passive neutron emission rate, measured fissile mass and the cooling time and initial enrichment.

The HRGS measurement is used to determine the activity of measurable gamma emitting fission products and activation products present in the hulls. ^{134}Cs, ^{137}Cs and ^{154}Eu fission product retention rates in the hulls batches are calculated from the measured isotope activities and the fission product specific activities determined from Feed Pond Fuel Monitor measured values of the fuel burnup, cooling time and initial enrichment. The retention rates are used to provide a diverse indication of gross dissolver maloperation in addition to that from the DDA measurement.

The satisfactory status of the Hulls Monitor system is demonstrated prior to every measurement by carrying out system standardisation checks. These comprise the measurement of background contamination in the empty hulls measurement thimble, a check of the satisfactory operation of the neutron and HRGS detectors and associated signal processing electronics by measurement of sealed sources automatically exposed under computer control and finally a check of the output of the neutron generator.

In view of the International Safeguards interest in fuel losses within the hulls waste, an independent data logging system extracts raw and processed data from the Hulls Monitor to permit checking and validation of its operation by Safeguards inspectors.

Commissioning Experience

An extensive range of commissioning measurements were carried out using

(i) Unirradiated UO_2 fissile standards,
(ii) Sealed ^{60}Co, ^{137}Cs and ^{252}Cf sources,
(iii) Irradiated UO_2 fuel standards.

The unirradiated fissile standards and sealed sources were used to characterise and calibrate the measurement systems during inactive commissioning. This included assessments of the impact of a range of LWR zircaloy and AGR stainless steel matrices, the errors associated with radial and axial residual fuel distributions in the hulls and the effect of gadolinium concentration and end appendages within the hulls matrix. The calibrations were validated during active commissioning using the irradiated fuel standards.

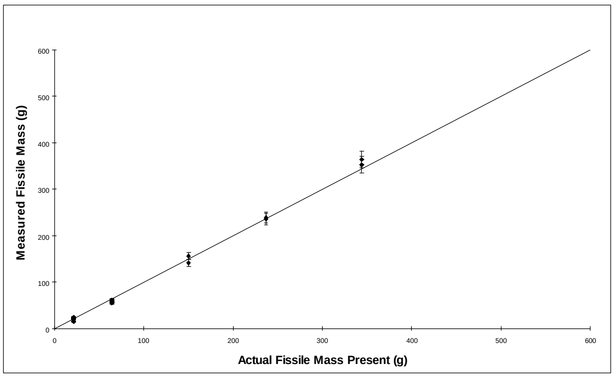

Figure 3 Hulls Monitor DDA Calibration Measurement Check

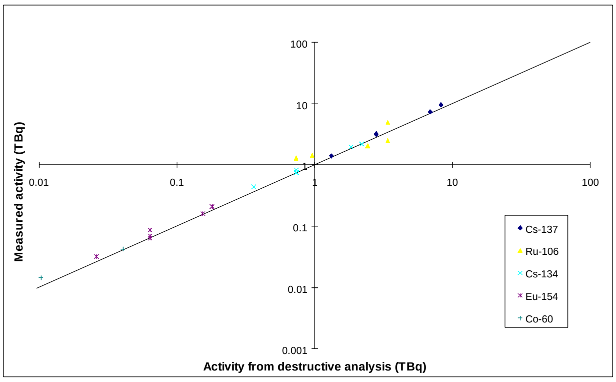

Figure 4 Gamma System Results Measuring Irradiated Fuel Standards

A plot of measured fissile mass against known mass values from fissile standards is shown in figure 3.

The random or statistical error on the measured fissile mass depends on the residual fuel carryover into the hulls and the irradiation history of the fuel. For example a recently reprocessed 31GWd/tU burnup PWR batch with a residual 1.8kg uranium carryover had a 38.8 ± 2.0 (1 σ statistical error) g^{235}U equivalent residual fissile content.

The detection limit for fissile mass is typically 2g ^{235}U equivalent fissile in a 600kg batch of leached BWR or PWR hulls.

A plot of measured gamma emitting fission product and activation product activities from the irradiated fuel standards against known radionuclide activities (determined from destructive analysis of samples of the fuel used to fabricate the standards), is shown in figure 4.

The operation of the Hulls Monitor was demonstrated during commissioning to Euratom Safeguards inspectors. This involved the independent verification of the fissile content of the Hulls Monitor unirradiated UO_2 fissile standards using a Euratom owned and operated Active Well Coincidence Counter and the subsequent “blind” measurement of a range of these unirradiated UO_2 fissile standards in measurement configurations specified by Euratom inspectors.

Operational Experience

The first active hulls batches were monitored in June 1994. Subsequently hulls from more than 6000 fuel assemblies have been successfully measured. This has demonstrated both the efficient operation of the THORP shear leach process and the performance of the Hulls Monitor.

During the reprocessing of the initial hulls batches in THORP, repeat measurements were carried out to characterise the performance of the Hulls Monitor. These included,

(i) Repeat measurements on hulls batches to confirm the random or statistical errors.

(ii) The measurement of hulls batches using both installed and spare neutron generators to demonstrate the absence of any systematic bias in their responses.

(iii) The physical redistribution of hulls matrices through the reorientation of hulls batches to investigate further and to confirm systematic errors associated with material positioning in a hulls batch.

(iv) Regular monitoring of emptied dissolver baskets to check for any residual background contamination.

(v) The regular measurement of “standard matrices” containing sealed sources, unirradiated and irradiated fuel standards in simulate hulls matrices to demonstrate the long term stability of the Hulls Monitor system.

The measurement of BWR hulls waste has demonstrated the preference for direct measurement of the uranium content using the active and passive neutron techniques as opposed to inference of the uranium content from the HRGS measurement. Figure 5 demonstrates for some BWR batches a significantly higher ^{137}Cs retention rate compared with the residual uranium retention rate. Any estimate of residual uranium mass based on the measured ^{137}Cs activity would result in a significant overestimate. The mechanism for the high and variable ^{137}Cs fission product retention is believed to be ^{137}Cs migration and impaction into the cladding during fuel irradiation. The large errors on the calculated ^{137}Cs retention rate for some batches are a result of variations in the burnup and cooling time characteristics of the nine BWR fuel elements in each dissolver batch.

Future Developments

BNFL Instruments are currently developing an Advanced Fuels Hulls Monitor to meet the additional challenges posed by high burnup (greater than 40GWd/t) UO_2 fuels and Mixed Oxide fuels with their significantly higher ^{244}Cm spontaneous fission neutron emissions. This system will replace the deuterium-tritium neutron generator with a pulsed superconducting Cyclotron of the type shown in figure 6. The cyclotron shown is being used in the BNFL Instruments neutron interrogation development rig which is currently being used to develop the Advanced Fuels Hulls Monitor. In addition, work is programmed to optimise the measurement technology for historical hulls waste which has been compacted for final disposal.

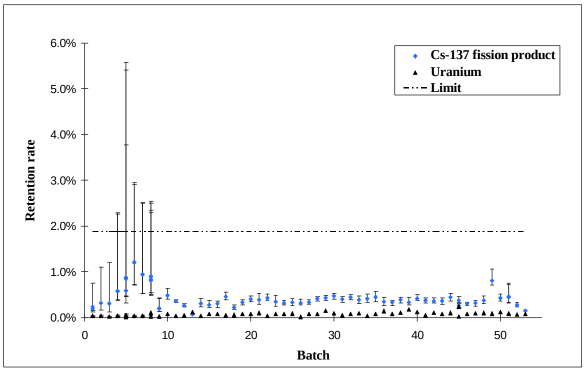

Figure 5 Residual Fuel and ^{137}Cs Retention in BWR Hulls Batches.

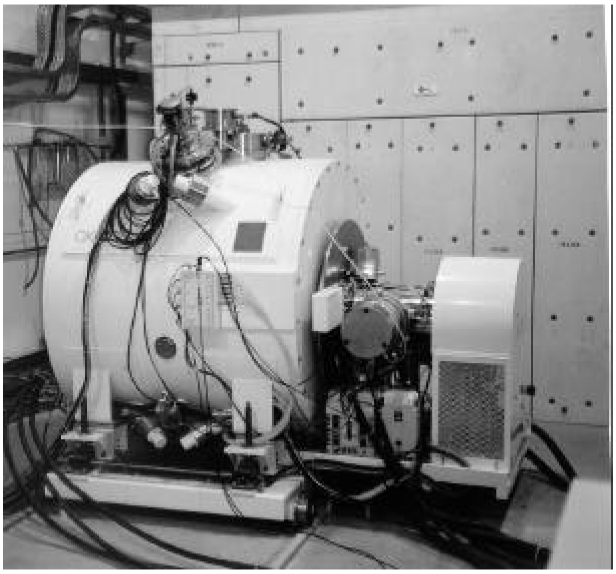

Figure 6 Radiometric Development Facility pulsed superconducting cyclotron.

CASE EXAMPLE 2: PLUTONIUM INVENTORY MEASUREMENT SYSTEM (PIMS)

Background

The Plutonium Inventory Measurement System (PIMS) is capable of determining the dynamic quantity and distribution of plutonium process materials throughout an operating plant. At plant clean out the same system will also show up residual material within the facility. PIMS is integrated into the plant providing data direct to the operators through the plant control computer systems. The PIMS installed within the THORP Product Finishing Line (TPFL) is also of interest for Safeguards, with mass data automatically reproduced at a Euratom monitoring station.

TPFL converts plutonium nitrate solution to plutonium oxide powder for storage and eventual use as MOX Fuel. There is a requirement for a measurement of the mass and distribution of plutonium throughout the plant covering all areas of significant inventory. Normal fissile material accountancy arrangements monitor the input and output of material throughout all of the operating plants at Sellafield. Any material remaining in the plant is assumed to be the difference between the material fed into the plant and product fed out. PIMS provides a near real time independent means of verifying this “inventory difference” book accountancy figure.

In addition, PIMS provides an early indication of abnormal build up or reduction of process material in certain key areas during normal plant operations. This measurement is required to provide process control as well as a TPFL inventory estimate for input into the THORP Near Real Time Material Accountancy System (NRTMA).

System Design

Commissioning and operational feedback has provided detailed knowledge of PIMS operation and performance both on THORP and in earlier applications. This feedback has enabled a continuous improvement in system design and operation, ensuring that future generations of PIMS are simpler to operate with improved performance and reliability.

PIMS systems are currently installed within three plutonium finishing lines on the Sellafield complex. A further system is being designed for installation into the Sellafield Mixed Oxide Fuel Fabrication Plant (SMP) which is under construction.

Figure 7 shows a block diagram of PIMS. This comprises neutron proportional counters, generally mounted within moderator modules, positioned at strategic locations throughout the plant. The system measures the total neutron count rate and mathematically unfolds these count rate responses to calculate the neutron emission from each process area. The neutron detector modules are positioned either singly or in arrays surrounding the process vessels of interest. Pulses from each detector are processed by a dedicated head amplifier. The data processing electronics comprise a number of standard commercially available electronics modules in addition to proprietary electronics systems.

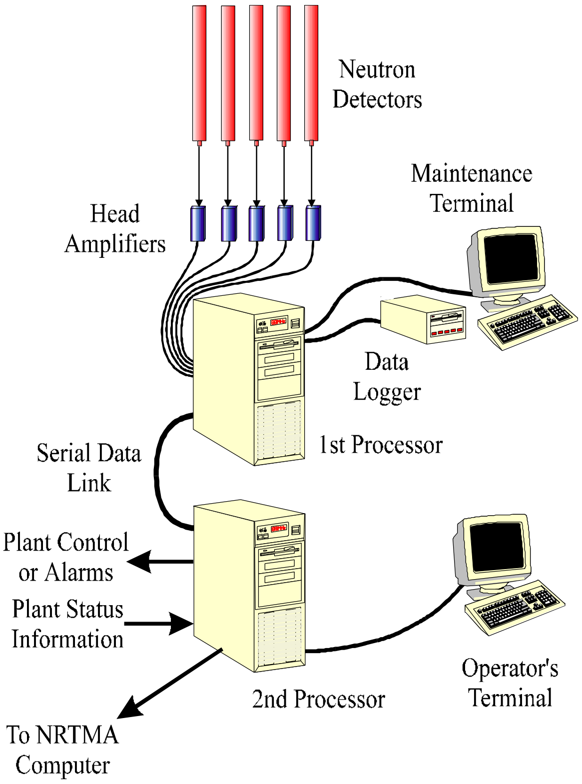

Figure 7 PIMS Block Diagram

Achieving accuracy in PIMS operation relies on the optimum positioning of the detectors relative to the plant process line. Typically, PIMS comprises between 30 and 100 detector modules positioned around the process vessels and gloveboxes throughout the plant. Each vessel/glovebox is modelled, either experimentally or using Monte Carlo neutronics codes to determine the optimum locations of the neutron detectors. The criteria for siting the detectors are:

- to maximise the detection efficiency of the detector (or array) to the process area being monitored;
- to minimise any system response variation over the process area being monitored due to different source positions;
- to minimise the detection efficiency of the detector (array) to adjacent areas of plant;
- to avoid dead-time problems due to excessive count rates.

In order to meet these criteria it is essential that PIMS is integrated into the plant design at an early stage, enabling the optimum detector locations to be achieved.

Figure 8 shows a simplified sketch of a plutonium plant layout including the main process vessels, and identifies the typical positions of the neutron detectors for PIMS.

The optimum detector module design has been determined using both computer based and experimental modelling work to minimise sensitivity to neutrons from adjacent process areas.

Calibration

Calibration involves moving an isotopic neutron source, usually ^{252}Cf, throughout the plant process line. The sensitivity of each detector to this “point” source in each of the calibration positions is accurately measured and used to build a calibration matrix, defining the response of each detector to neutron emitting material within each process area being monitored. The calibration matrix is used to interpret the count rate data from the detectors and to estimate the neutron emission, and hence the plutonium mass, in each of the areas. The matrix solution technique uses an iterative weighted least squares technique to estimate the plutonium mass (and its associated measurement uncertainty) in each process area.

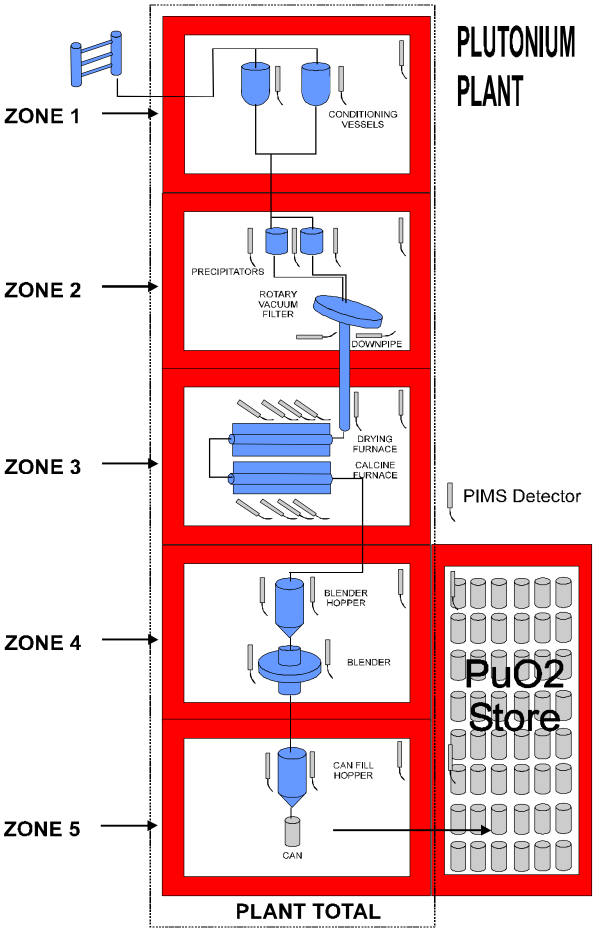

Figure 8 Typical PIMS Detector Locations on a Plutonium Finishing Facility

Operational Experience

As well as the experience from application on earlier Sellafield plutonium plants, the THORP PIMS has been successfully in operation since TPFL started processing active material and provides three levels of results: the overall plant inventory total, the individual masses of the active cells, plus process control figures for some of the main gloveboxes and vessels.

PIMS provides both plant operators and regulators with confidence and knowledge of the way in which the plant is operating. This is illustrated by the following examples.

Figure 9 shows the PIMS mass results for the plant feed glovebox during the first commissioning feed of plutonium. The plant feed glovebox conditions and dilutes the plutonium nitrate liquor at the start of the process. The initial build up of plutonium can be easily seen following the start up. The dip at run hour 100 is due to an interruption in the feed. The reported mass for this glovebox when compared with an estimated figure based on throughput is within ± 5% of the actual inventory.

Figure 10 shows the mass results down stream for the Vacuum-Separator vessel. The oxalate mother liquor filtrate from the filter bed is removed through this vessel, and this vessel should contain minimal Pu in normal operation. The break through of material through the filter table can be seen by the plant operator and remedial action taken.

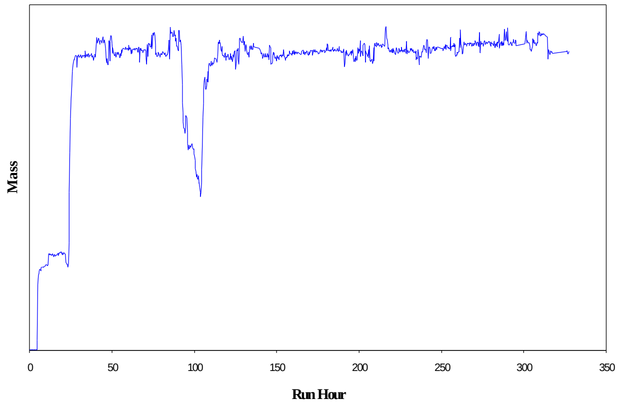

Figure 9 Plant Feed Glovebox Results

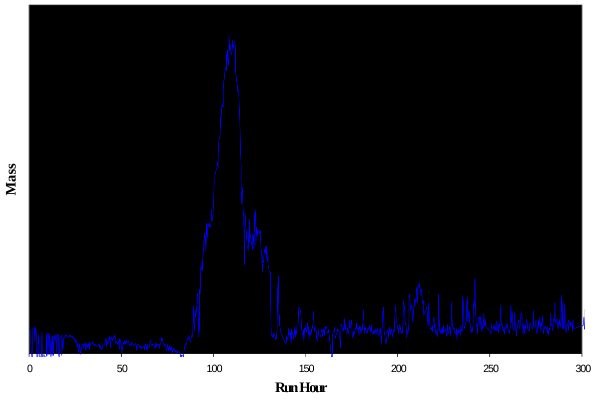

Figure 10 Vacuum-Separator Vessel Results

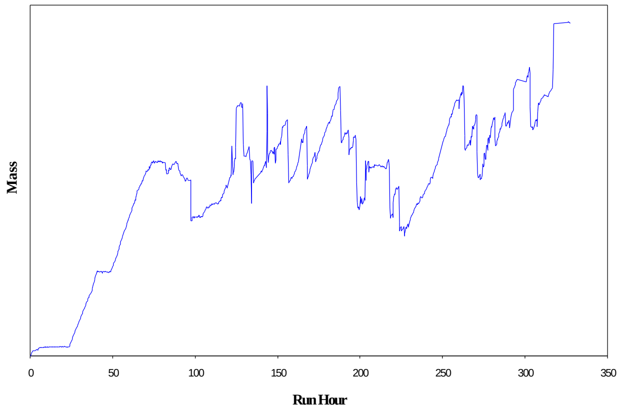

Figure 11 PIMS Total Plant Mass Estimates

Figure 11 shows the mass results for the total plant inventory. The build up of mass as material feeds through the plant and the effect of removal of cans of material from the total inventory of the process line to the adjacent store can be seen as the saw tooth effect from run hour 100 onwards. This result feeds directly to the NRTMA system providing an estimate of the plant inventory on a near real-time basis, forming part of the THORP Safeguards approach.

Typically the PIMS agrees to within 5% of the actual plant inventory.

Future Developments

A commercially available version of PIMS known as FissTrack™ is under product development. In addition on-going development of the PIMS technology includes overcoming problems with neutron multiplication effects. This arises when PIMS technology is applied to large batch size processes such as used for MOX fuel fabrication.

CONCLUSIONS

The control of operations in spent nuclear fuel recycle plants, and the monitoring of special nuclear material within those plants, gives rise to a variety of requirements for measurement of radionuclides. Many of these can only be satisfactorily met by the provision of on-line special instrumentation.

The cost of provision of such special instrumentation must be justified by the benefits resulting from their use. The close and continuing dialogue between the instrument supplier, the plant designer, the safeguards authorities and the plant operator, is invaluable in maximising this benefit.

Given that, by definition, these instruments are located at key points in the process, particular attention must be paid to the design and construction of such special instruments to maximise their reliability and minimise down time.

The THORP project represented a major exercise in special instrument development which has now been successfully completed with demonstrable benefits to the operation and safeguarding of the facility.

The instruments highlighted in this paper have achieved their design intent. Complex measurements are achieved with an accuracy which has allowed the operator to gain a detailed insight into the operation of his process. The experience gained in their commissioning is already proving beneficial in extending the technologies to the fabrication and future reprocessing of mixed oxide fuels.