IMPROVED ASSAY OF HIGHLY ACTIVE REPROCESSING WASTE BY THE USE OF A LINEAR ACCELERATOR BASED NEUTRON SOURCE

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Abstract

Dissolution of spent fuels in the THORP reprocessing plant results in large batches of spent fuel cladding which must be comprehensively monitored prior to its export from the plant. The Hulls Monitor has operated successfully since THORP went on stream in 1994, providing vital data for process control, criticality safety, nuclear material accountancy/safeguards, and inventory determination prior to ultimate disposal. The system utilizes a combination of three measurement techniques: active neutron Differential Die-Away, passive neutron counting and high resolution gamma spectrometry. The Differential Die-Away measurement on fuel residues in an industrial environment demands a commercially available, robust, pulsed high output neutron source. The original D-T generator delivered a detection limit of 2g^{235}U equivalent in 600kg of zircalloy cladding. However current operational issues and future trends in the fuel to be reprocessed have provided the stimulus to develop alternatives. This paper describes the incorporation of a compact AccSys DL-1 linear accelerator neutron source in the Hulls Monitor and its successful commissioning and operation in THORP. The next generation of Hulls Monitor systems under development is also described. This will address the measurement challenges posed by the reprocessing of mixed plutonium / uranium-oxide fuels and very high burnup uranium oxide fuels.

Introduction

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. The THORP Hulls Monitor is one of over thirty types of such special instrument systems that have been developed and commissioned during the 1980s and early 1990s on the THORP commercial reprocessing plant at Sellafield.

The Hulls Monitor is installed in the THORP Head End plant where oxide fuels are removed from their storage flasks, monitored by the Feed Pond Fuel Monitor [i ii], sheared into 1.6te dissolver batches and dissolved for subsequent chemical separation. 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. Typically 600kg of zircalloy or stainless steel hulls from each dissolver batch are measured in a dissolver basket, which has a diameter 700mm and maximum fill height of 2300mm. The Hulls Monitor measurements are required:

1. To assure criticality safety of the hulls during subsequent handling in THORP and the Waste Encapsulation Plant.
2. To provide process control data on the leach efficiency to permit sentencing of each hulls batch.

3. 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.
4. To provide an activity inventory for compliance with Intermediate Level Waste contractual agreements and repository acceptance criteria.

This paper describes the Hulls Monitor and how current operational issues and future trends in the fuel to be reprocessed have provided the stimulus to incorporate a compact AccSys DL-1 linear accelerator based neutron source in the Hulls Monitor. Also described is the next generation of Hulls Monitor systems under development at Sellafield which will address the measurement challenges posed by the reprocessing of mixed plutonium / uranium-oxide fuels and very high burnup uranium oxide fuels in later years of THORP's operation.

Hulls Monitor System and Technique

The Hulls Monitor (see Figure 1) 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. The passive neutron measurement is used as an input to determine safeguards and inventory results. The HRGS measurement gives the radionuclide inventory of the high energy 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 Sodern Genie-26 deuterium-tritium 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 or absorbed. The resultant thermal neutron flux decays over a period of hundreds of micro seconds and will induce fissions in any fissile material present producing "secondary" fast neutrons. It is these secondary fast neutrons which are counted to give a measure of the fissile material present. Figure 2 shows examples of fast neutron flux decay in irradiated fuel hulls batches containing 3g and 250g ^{235}U equivalent residual fissile content.

The passive neutron emission rate, comprising primarily spontaneous fission neutrons from ^{244}Cm is measured as the dissolver basket is raised through the moderating collar. The passive

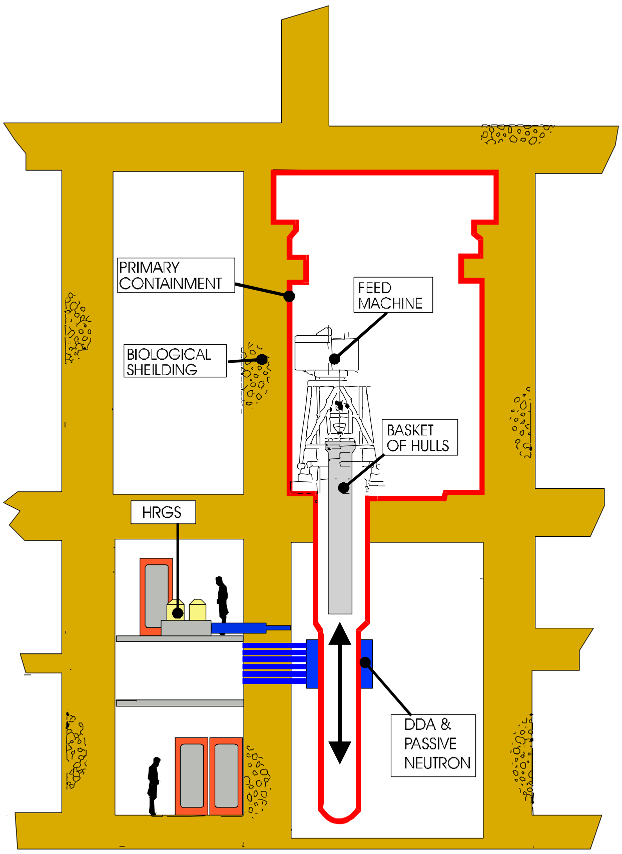

Figure 1 Schematic Arrangement of the Hulls Monitor.

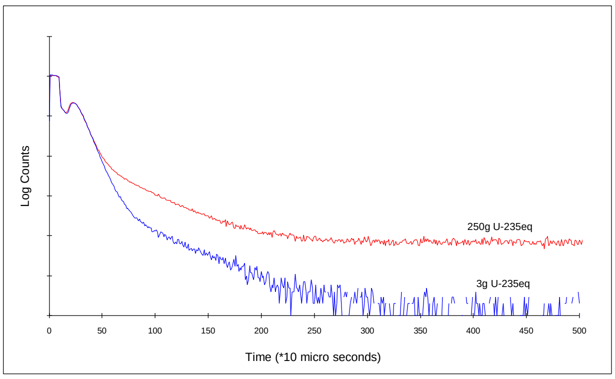

Figure 2 Decay of Fast Neutron Flux in Hulls Monitor DDA Measurement Cavity

neutron emission rate then is used with the measured fissile mass and the initial enrichment, from the Feed Pond Fuel Monitor measurement before shearing, to calculate 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 calculated from the measured passive neutron emission rate, measured fissile mass and the cooling time and initial enrichment, from the Feed Pond Fuel Monitor measurement before shearing.

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, which are determined from the Feed Pond Fuel Monitor measured 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. Finally the output of the neutron generator is checked.

In view of the International Safeguards interest in tracking fuel retained within the hulls waste as a possible diversion route, an independent Euratom data logging system extracts raw and processed data from the Hulls Monitor to permit checking and validation of its operation by Safeguards inspectors.

<u>Hulls Monitor Commissioning and Operational Experience</u>

Well characterized unirradiated UO_2 fissile standards and sealed ^{60}Co, ^{137}Cs and ^{252}Cf sources have been used to extensively characterize and calibrate the measurement systems during inactive commissioning. This included assessments of the impact of a range of LWR zircalloy and AGR stainless steel matrices, the total measurement uncertainty 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 well characterized irradiated UO_2 fuel standards. A plot of the measured fissile mass against known fissile masses of fissile standards is shown in Figure 3. The total measurement uncertainty calculated by the Hulls Monitor, shown on Figure 3, is the total measurement uncertainty for the system's criticality safety measurement.

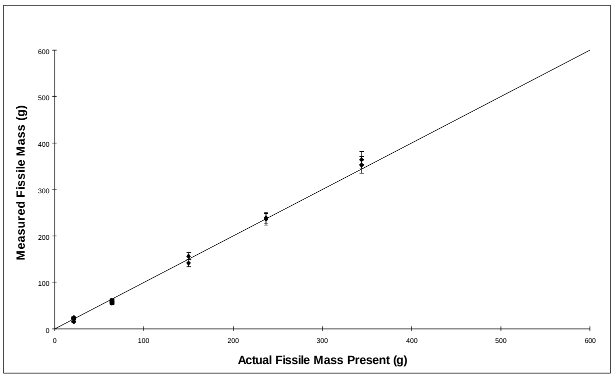

Figure 3 Hulls Monitor DDA Measurement Calibration Check

The random or statistical uncertainty on the measured fissile mass depends on the residual fuel carryover into the hulls and the irradiation history of the fuel. For example a 31GWd/tU burnup PWR batch with a residual 1.8kg uranium carryover was measured as 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. The most significant source of systematic uncertainty arises from geometrical variations in the location of the residual fuel within the hulls basket. For criticality safety, the upper uncertainty on the measured result is calculated assuming a worst case fuel distribution, to determine the highest possible fissile mass that gave rise to the measured signal. A less pessimistic assumption of the fuel distribution is used to determine the negative systematic uncertainty on a measured result.

The operation of the Hulls Monitor was satisfactorily demonstrated during commissioning to Euratom Safeguards inspectors. This involved a series of "blind" Hulls Monitor measurements of the unirradiated UO_2 fissile standards in measurement configurations specified by Euratom inspectors. As part of this exercise, Euratom carried out an independent confirmation of the fissile content of the Hulls Monitor unirradiated UO_2 fissile standards using an Euratom owned and operated Active Well Coincidence Counter.

The first active hulls batches were monitored in June 1994. Subsequently hulls from more than 2000 tonnes of fuel in approximately 1300 hulls batches 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 BWR, PWR and AGR hulls batches in THORP, repeat measurements were carried out to demonstrate the reliability of the Hulls Monitor and characterize its performance. These included,

1. Repeat measurements on hulls batches to confirm the random or statistical uncertainties.
2. The measurement of hulls batches using both installed and spare neutron generators to demonstrate the absence of any systematic bias in their responses.
3. 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.
4. Regular monitoring of emptied dissolver baskets to check for any residual background contamination.
5. 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 supports the choice of a direct DDA 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 4 illustrates for some BWR batches a significantly higher percentage of the initial ^{137}Cs in the fuel (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 fuel mass 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

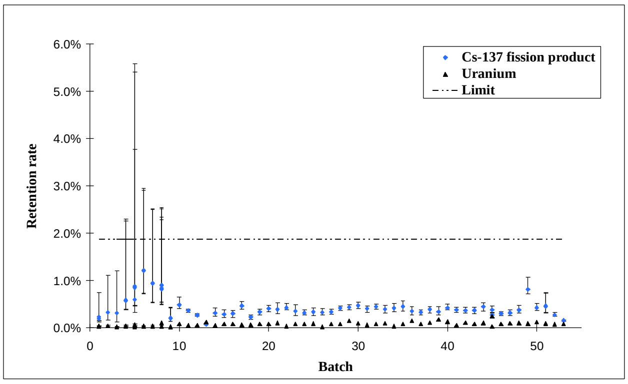

Figure 4 Residual Fuel and ^{137}Cs Retention in BWR Hulls Batches.

fuel irradiation. The large uncertainties shown 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.

Operational experience with the Sodern Genie-26 deuterium-tritium neutron generators showed that they require routine preventative maintenance and that their neutron output decreases significantly with operation as shown in Figure 5. This necessitated the routine replacement of the generator's neutron emitting probe unit.

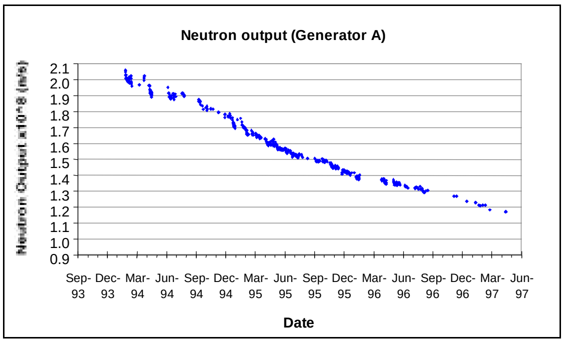

Figure 5 Reduction of Sodern Genie-26 deuterium-tritium neutron generator output with time.

The Hulls Monitor application of the DDA technique requires a high output neutron source providing a minimum depending on the fuel characteristics, of 1.5×10^8 neutrons per second at a frequency of 15 pulses a second. This high neutron output is at the current limit of technology for deuterium-tritium neutron generators, together with reliability and security of supply concerns led BNFL to consider alternative neutron sources.

Following an evaluation of commercially available systems the decision was made to install an AccSys DL-1 Linac as a replacement for one of the Hulls Monitor's two deuterium-tritium neutron generators.

<u>AccSys DL-1 Linac</u>

The AccSys DL-1 Linac[iii] is a Radio Frequency Quadrupole (RFQ) Linac that accelerates deuterium ions to 0.9 MeV. The Linac uses a $^{9}Be(d,n)^{10}B$ reaction in a beryllium target to produce the neutron output. The Linac's compact ruggedised design and intrinsically pulsed neutron output makes it a suitable replacement for a deuterium-tritium neutron generator tube in an industrial DDA application. The expected maximum output from a fully tuned system at the Hulls Monitor duty cycle of 15Hz is 1×10^9 neutrons per second. The system is currently operating with a maximum output of 5×10^8 neutrons per second, which is still significantly greater than can be achieved by any currently available deuterium-tritium neutron generator design.


Figure 6 AccSys Linac DL-1

The choice of the Linac as a replacement for one of the two deuterium-tritium neutron generators affords both diversity of supply and an increase in operational flexibility. Thereby allowing the higher output Linac to be used in the measurement of more challenging higher passive neutron emission hulls batches.

In order for the Linac to be installed it had first to be demonstrated that the system could be operate reliably and provide the neutron pulse structure of a deuterium-tritium neutron generator tube. This would allow the Linac to be installed with minimal effect on the calibration of the system allowing a rapid commissioning on THORP and minimizing any plant operational disruption. The operation of the Linac with the deuterium-tritium neutron generator pulse structure of 90µs pulses at 15 Hz for a 90 minute measurement duration was within the operational envelope of the system. However the 0.9 MeV $^{9}Be(d,n)^{10}B$ reaction produces a neutron energy spectrum from 0.25 MeV to 6 MeV with a peak output at approximately 1 MeV. This output is very different to the monoenergetic 14 MeV energy spectrum of the deuterium-tritium neutron generator. The effect of the different neutron output on the performance of the system was evaluated by modifying the benchmarked Hulls Monitor MCNP[iv] model.

Table 1 shows that the higher energy 14MeV neutrons from the deuterium-tritium neutron generator interact with the lead collar of the chamber, shown in blue in Figure 7, producing an additional component of neutron flux via (n,2n) reactions in lead. The lead is used to shield the neutron detectors from the intense gamma radiation from the hulls basket. There are no (n,2n) reactions with the Linac generated neutrons as these are all below the energy threshold for this reaction. This gain in neutrons for the deuterium-tritium neutron generator is partially offset by additional losses of neutrons escaping into the surrounding concrete and fast neutrons absorbed inside the Hulls Monitor mainly due to $^{12}C(n,a)$ reactions in the graphite and polythene moderating materials in the measurement chamber. This interaction has a threshold at 6 MeV.

	D-T	Linac
Neutrons created		
Emitted by generator	100	100
Extras from (n,2n)	40	0
Total	140	100
Neutrons killed		
Thermal Neutron absorption	125	96
Fast neutron absorption	4	<0.1
Escape into concrete	11	4
Total	140	100

Table 1 MCNP creation and loss of neutrons (per 100 neutrons generated

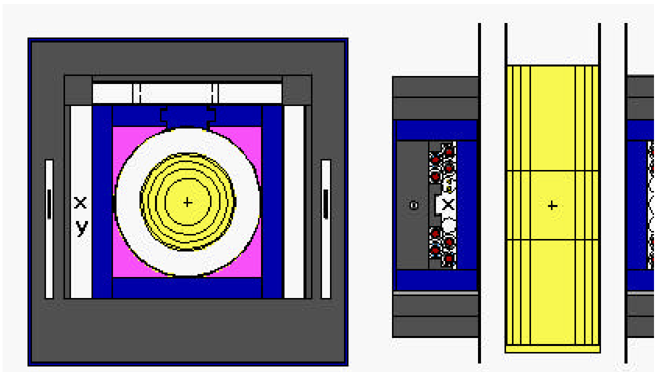

Figure 7 MCNP model of the Hulls Monitor.

For a given neutron output, the response that the model predicted for each generator was found to be similar. However the lack of the (n,2n) derived component of flux from the Linac meant that the resulting thermal flux within the measurement chamber was slightly reduced and less axially distributed than that of the deuterium-tritium neutron source. These differences necessitated a level of recalibration of the system.

In order for the Linac to be installed in to the Hulls Monitor a longer beam line was designed and constructed to ensure that the neutron producing target was at the same position as the deuterium-tritium neutron generator target (marked X in Figure 7). This high energy beam transport line required electromagnetic steering and focussing elements to ensure that the ion beam could be accurately aligned on the target section.

The Linac system was extensively tested during a series of factory acceptance tests. These tests evaluated the structure of the pulsed output of the system by measuring the beam current incident on the target section. The neutron output of the Linac remained constant to within a few percent over the ninety minutes of each measurement run, producing over 99.5% of the expected pulses at the full output level.

The AccSys Linac was installed on THORP in August 1998. After a set to work and brief commissioning period the performance of the Hulls Monitor with the Linac as the neutron source was evaluated. The calibration parameters were modified to account for the difference in the neutron spectrum predicted by the MCNP modelling by the measurement of fissile standards. Following these modifications the Linac based measurement system was demonstrated to produce results consistent with the alternative deuterium-tritium neutron generator system for a range of fissile standard and real hulls measurements.

The Linac currently operates in a "manual mode" for the measurement and sentencing of hulls batches. The system will be fully integrated into the Hulls Monitor control systems when the necessary Linac control software has been implemented. The new system providing the diversity, security of supply and potential for increased neutron for future operational flexibility.

Advanced Fuels Challenges

BNFL Instruments are currently developing an Advanced Fuels Hulls Monitor to meet the additional challenges posed by very high burnup (significantly greater than 40GWd/t) UO_2 fuels and Mixed Oxide "Advanced Fuels" with their significantly higher spontaneous fission emissions from ^{244}Cm.

This development work is being carried out in BNFL Instruments purpose built Radiometric Development Facility shown in Figure 8. This comprises a shielded cell providing a large experimental area for the development of such large scale interrogation systems and contains several experimental rigs and radiation generating devices used to develop and test a range of radiometric instrument systems.

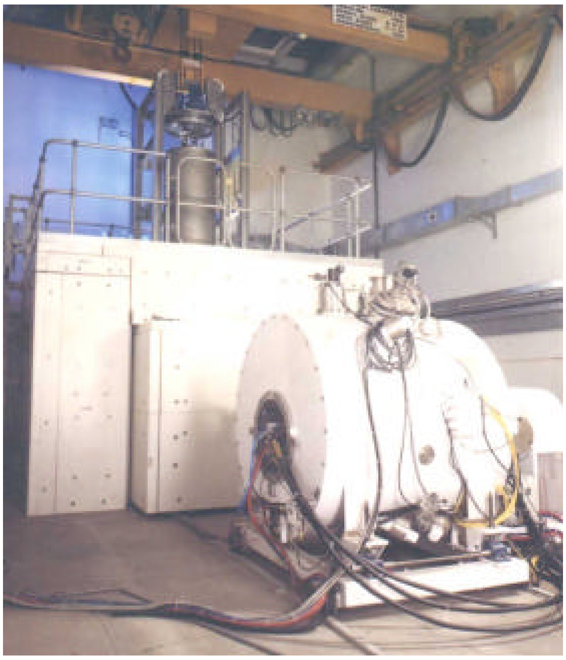

Figure 8 Radiometric Development Facility pulsed superconducting cyclotron and Advanced Fuels Hulls Monitor development rig.

The higher passive neutron emission from "Advanced Fuels" leads to a requirement for a larger neutron source in order to measure the fissile content, effectively a consequence of signal to noise ratio. The Advanced Fuels Hulls Monitor is being developed using a superconducting Cyclotron of the type shown in Figure 8 as an intense neutron source. This system is a development by Oxford Instruments' of their OSCAR 12 MeV proton cyclotron usually employed in isotope generation. With the addition of a beryllium target, the system is capable of producing a continuous output of $2x10^{12}$ neutrons per second. For the Advanced Fuels Hulls Monitor application, the system was modified to produce a wide range of pulsed duty cycles and neutron outputs, for supporting both delayed and prompt neutron measurement modes. When operating at the Hulls Monitor duty cycle the output is $2x10^9$ neutrons per second.

In addition, the development of the Advanced Fuels Hulls Monitor has led to a number of additional improvements over the Hulls Monitor existing system.

1. Using a combination of MCNP modelling and experimental measurements, the mechanical configuration of the system has been optimized to maximize the induced fission signal. This led

to the installation of the neutron-generating target at the center of a moderating assembly, located externally to the measurement chamber.

2. The Advanced Fuels Hulls Monitor system will use two types of neutron detector. This dual approach allows the low efficiency detector set to operate in high background conditions that will saturate the response of the main ^{3}He detector set. The low efficiency detector set, which is based on ^{4}He detectors, widens the dynamic range of the measurement, allowing with the most challenging fuel batches to be monitored (i.e. high fuel carry-over and high background emissions).
3. Modifications to the method of hulls matrix identification and correction have been developed to improve the accuracy of the system and reduce the uncertainty in the results. The method of data collection and analysis has been enhanced to allow study of the structure of the interrogating flux and fissile response. The variation in the fissile response of the system as the basket scans helically through the measurement chamber is used in an imaging algorithm to determine the distribution of fissile material within the hulls. Complementary measurement of the interrogating flux using flux monitor detectors measure the absorption properties of the Hulls batch showing local variations due to items such as PWR end appendages. Therefore, corrections for the position and distribution of the fissile material can be made, reducing the systematic uncertainty in the measurement results.
4. The direct measurement of the neutron absorption and moderation properties of the hulls matrix removes the requirement for changing calibration parameters at the start of each new fuel campaign. This reduces the uncertainty arising due to neutronic differences between the measured hulls matrix and the calibration matrix.

Conclusions

The Hulls Monitor provides a very sensitive and accurate measurement of leached hulls batches in a very challenging high radiation environment. The Hulls Monitor generates vital data for process control, criticality safety, nuclear material accountancy, international safeguards, and inventory determination purposes.

The successful use of the accelerator based neutron sources in Hulls Monitor systems have been demonstrated. The AccSys DL-1 Linac has provided a reliable intense neutron source for the THORP Hulls Monitor DDA measurement providing a diversity of supply and greater operational flexibility. In addition a pulsed neutron producing superconducting cyclotron has been successfully used for the development of an Advanced Fuels Hulls Monitor in the BNFL Instruments Radiometric Development Facility.

References

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