Application of the PAN/GEA/AK method to the Non-Destructive Assay of Remote Handled TRU Waste.

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ABSTRACT

Combining Passive Active Neutron / Gamma Energy Analysis with process or acceptable knowledge (the PAN/GEA/AK technique) has been successfully deployed for the characterization of contact handled (CH) transuranic (TRU) waste in accordance with the regulatory requirements. For most CH TRU streams Pu240 is the dominant source of spontaneous neutron emission, Pu239 is the dominant fissile isotope. Thus the signal from passive coincidence techniques may be attributed to Pu240 and the active neutron signal may be attributed to Pu239. Gamma spectroscopy (or AK data where insufficient gamma data is available) is then used to discriminate the other nuclides contribution to the active and passive signals.

For Remote Handled TRU (defined as containers with external dose rates in the range 0.2 to 1,000 R/hr) it is often mistakenly believed that active and passive neutron techniques are not appropriate, because (i) isotopics are difficult to determine by gamma spectroscopy (ii) the waste streams are more diverse with isotopes such as the spontaneous neutron emitter Cm244 present for which there is no means to discriminate against. Both of these points are undeniably true – isotopics measurements are compromised by the Compton background and there is no easy means to discriminate different spontaneous neutron emitters or different fissile isotopes based purely on the signals received by the NDA equipment. However, BNFL Instruments experience gained from operations in the UK and USA using PAN/GEA/AK techniques to assay arisings from diverse waste streams including spent fuel debris (metal fuels and oxide fuels), reprocessing facility streams (waste and process measurements), plutonium fluoride, heat source plutonium (Pu238), isotope production facility waste (Cm244, Cm246, Cf252 etc), weapons facility waste, uranium waste (various enrichments from depleted to HEU) has shown that regulatory acceptable measurements can be made under the most challenging conditions. Expert analysis may be used in combination with automated software routines to make intelligent, experience based decisions on the waste container radionuclide inventory even when presented with "problem waste streams" producing signals that may, at first, be regarded as intractable. During design and commissioning of the NDA equipment, algorithms can be established to deconvolve the data for the known waste streams and pre-program data quality flags that indicate containers whose assay results fall outside the valid range of these algorithms. For the outlier containers that are thus flagged, the expert will make use of acceptable knowledge, sampling data together with all available assay information to produce defensible estimates of the radiological contents of the container.

INTRODUCTION

The characterization of transuranic waste by combining information from multiple NDA techniques and from acceptable knowledge (AK) is a mature approach that has evolved to comply with increasingly stringent regulatory requirements for a period of more than 20 years. The processes for combining the information from multiple measurement techniques with AK have become quite complex, relying on rule-based decision-making schemes and mathematical formulae to provide defensible regulatory compliant characterization results for a wide variety of waste streams. To the extent practicable, these schemes and formulae are applied by automated software routines to provide characterization results data. The automated software has also been developed to recognize exception conditions, where expert analysis is required. In such cases the software provides pre-programmed flag conditions, which serve to disposition the analyses to an expert analyst.

The combination of Passive/Active Neutron measurements, Gamma Energy Analysis and acceptable knowledge (PAN/GEA/AK technique) is an example of a methodology that combines information from multiple NDA techniques with AK to provide TRU characterization. The PAN/GEA/AK technique has matured and evolved in conjunction with its application to a variety of waste streams and the evolution of stringent regulatory requirements.

While a more simplistic approach to the characterization of Remote Handled (RH) TRU could be desirable from the standpoint of cost and time, RH TRU streams present significant challenges to NDA techniques and it is unlikely that simplistic characterization schemes will be capable of meeting these challenges and complying with anticipated regulatory requirements.

The experience gained from applications of this technique to RH streams in the UK have demonstrated that the technique itself requires careful tailoring to overcome the challenges presented by specific waste streams and regulatory requirements.

OVERVIEW OF THE PAN/GEA/AK TECHNIQUE

The PAN/GEA/AK technique combines three distinct NDA measurements. Combining the data from these 3 resultant measurements and additional AK as required provides the final reportable characterization results. The NDA measurements consist of a passive neutron measurement, an active neutron measurement and a gamma energy analysis measurement.

Passive neutron counting consists of measuring the intrinsic fast neutron emission from the waste. The technique includes the detection of events involving two time-correlated neutrons (passive neutron coincidence counting PNCC) and the detection of multiple time-correlated neutrons (Multiplicity Counting). Passive neutron measurements are used to quantify even isotopes of plutonium and other spontaneous neutron emitters [1].

The active neutron measurement consists of measuring neutron-induced fission events in fissile material such as Pu239 within the waste. Neutrons from an interrogating source are introduced into a measurement chamber consisting of moderating and shielding materials. These neutrons induce fission events in fissile material, giving rise to the emission of secondary fast neutrons. Various techniques are used to maximize the sensitivity to secondary neutrons, while minimizing the signal from the interrogating source. One widely used method is the differential die-away (DDA) technique. Short pulses of fast neutrons from a neutron generator are injected into the measurement chamber. This gives rise to a thermal neutron flux, which persists for a few milliseconds. Fast neutrons arising from the induced fission events are then counted using fast neutron detector packages embedded in the chamber walls [1].

Gamma Energy Analysis consists of analyzing gamma spectra to quantify specific isotopes or to ascertain mass fractional data for those isotopes with characteristic photo peaks above the lower limit of detection. Quantification of gamma-emitting isotopes requires establishing an efficiency curve, which accurately corrects for the sample matrix attenuation of the photo peaks. The mass fractional data may be obtained by mathematical models, which establish a “relative efficiency curve” for the waste container by utilizing the branching ratios of isotope(s) with multiple peaks and the detector efficiency characteristics.

AK data consists of additional knowledge regarding the waste, that satisfy regulatory requirements for “acceptable” and that can be utilized to assist in the quantification of reportable radioisotopes within the waste. An example of AK would be information on the origin of the radio isotopic species in the waste that would allow the quantity of a specific isotope with no characteristic photo peaks to be correlated from the measured quantity of other isotopes in the waste.

The combining of the data from both neutron measurement techniques (PAN), the gamma analysis (GEA) and the AK data is a complex set of rules and mathematical formulae that are developed for specific waste streams with specific measurement conditions and specific regulatory requirements for characterization. Generally, the vast majority of the waste containers for a specific waste stream are characterized through automated software that incorporates the set of rules and mathematical formulae to produce reportable characterization data. In addition, the software checks for exception conditions and flags these containers for disposition to expert analysis. The expert analysis consists of utilizing experience to provide defensible methodology for characterizing the exception containers.

HISTORICAL APPLICATIONS OF THE PAN/GEA/AK TECHNIQUE

Specific historical applications of the PAN/GEA/AK technique illustrate the capabilities of this technique in a variety of regulatory environments to characterize diverse waste streams. The technique or variations of the technique have been utilized in the UK and the US.

Example 1: Thermal Oxide Reprocessing Plant (THORP) Hulls Monitor.

The THORP reprocessing plant at Sellafield in the UK receives and reprocesses several thousand tons of spent light water reactor (LWR) and advanced gas cooled reactor (AGR) fuel. After dissolution of sheared fuel, the resulting pieces of empty fuel cladding (hulls) are measured by the Hulls Monitor (figure 1) prior to encapsulation and eventual disposal. Hulls Monitor measurements are required to (i) assure criticality safety of the hulls during subsequent handling in THORP and the Waste Encapsulation Plant, (ii) provide process control data on the leach efficiency to permit sentencing of each hulls batch, (iii) determine the residual masses of U235, total plutonium and fissile plutonium in the hulls for materials accountancy and Safeguards purposes, (iv) provide an activity inventory for compliance with Intermediate Level Waste contractual agreements and anticipated repository acceptance criteria [2,3].

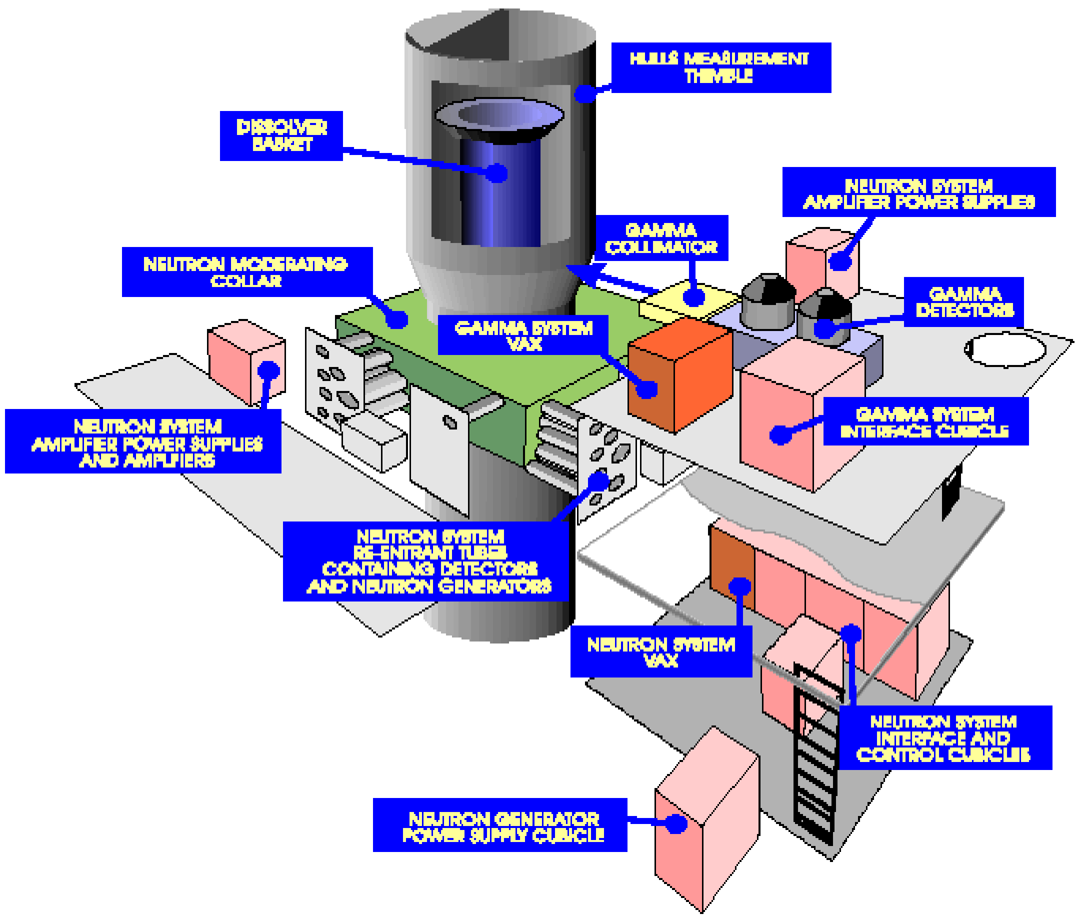

Fig. 1 Schematic of Hulls Monitor

The Hulls Monitor uses three radiometric techniques: Differential Die Away (DDA) active neutron measurement, passive neutron measurement and High Resolution Gamma Spectrometry or GEA. The DDA determines the residual fissile content of the hulls. The passive neutron emission rate is used in conjunction with the fissile mass from DDA and with the initial enrichment and cooling time (from measurements of the spent fuel prior to its shearing and dissolution in the Feed Pond Fuel Monitor to determine the residual uranium mass, the Cm244 activity, total Pu activity, total U activity, total alpha activity, U235 mass, fissile Pu mass and total Pu mass. The GEA measurement is used to determine the activity of measurable gamma emitting fission products and activation products present in the hulls [2].

The operation of the Hulls Monitor was demonstrated during commissioning to Euratom Safeguard inspectors. This involved the independent verification of the fissile content of the Hulls Monitor un-irradiated UO_2 fissile standards using a Euratom owned and operated Active Well Coincidence Counter and the subsequent "blind" measurement of a range of these fissile standards in measurement configurations specified by the inspectors. Additionally, during the reprocessing of the initial hulls batches in THORP, replicate measurements were conducted to characterize random errors, to demonstrate absence of systematic bias from neutron generator output, and to assess sources of total measurement uncertainty (TMU) such as distribution of hulls, residual background contamination and hull matrix effects. [2].

Example 2: Miscellaneous Beta Gamma Waste Store (MBGWS) Fissile Material Detector.

The first on-plant application of the DDA neutron interrogation technique at Sellafield was the Fissile Material Detector (FMD) at the MBGWS. This store receives general items of high activity waste from the reprocessing plants. These items are repackaged and stored in 3 m^3 boxes. The FMD measures the fissile content of the waste for criticality safety purposes prior to filling the boxes [1].

The measurement chamber consists of lead, polyethylene and graphite, with neutron detectors and a pulsed neutron generator located in the walls. Thick lead shielding (6") is required to shield the neutron detectors from the high gamma dose rates routinely encountered (~10's of Sv/hr). Automated checks on performance are carried out using a neutron source transfer system. The system is capable of measuring the wide variety of wastes consigned to the store, which are categorized as either filters, combustibles or non-combustibles prior to assay. Various calibrations were performed for each declared waste classification during commissioning including mixtures of steel, lead, concrete, graphite, cellulose and plastics. The total fissile content is derived using the DDA measurement signal, operator declared classification and measured neutronic properties of the waste consignment [1].

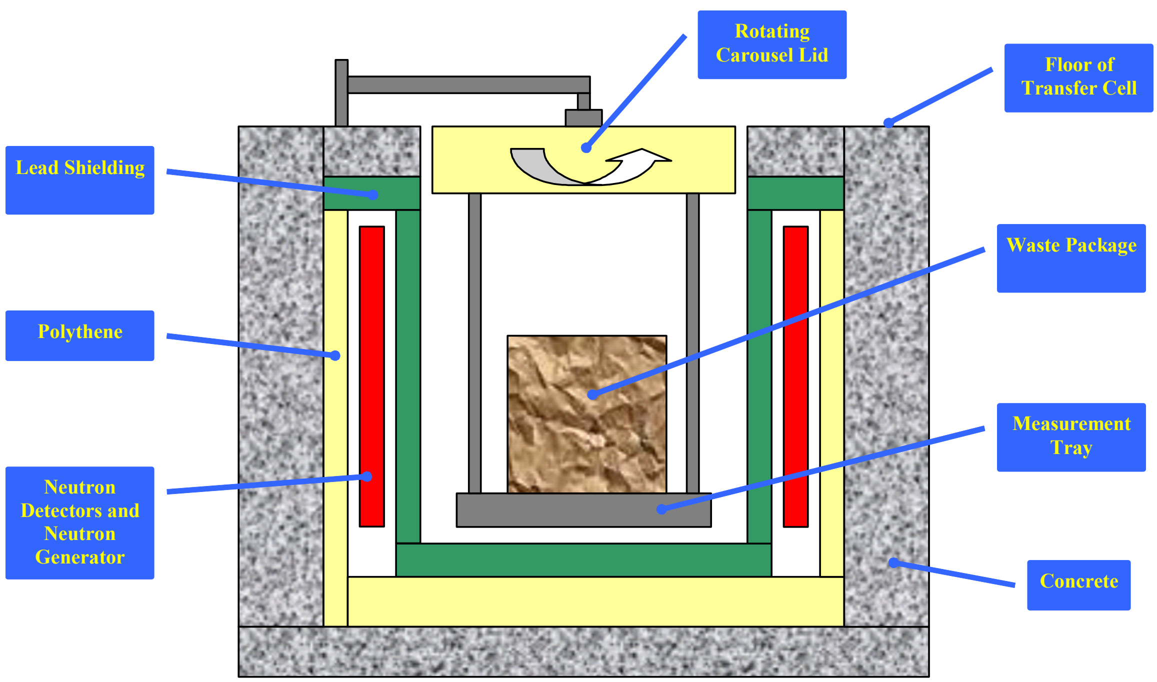

Fig. 2 Schematic of Fissile Material Detector

Example 3: Mobile Imaging Passive and Active Neutron (IPAN) System

The DDA technique has been expanded in the US to facilitate the performance of Waste Isolation Pilot Plant (WIPP) certified measurements on waste drums and waste crates. This technique is referred to as Imaging Passive and Active Neutron (IPAN™) transuranic assay. The passive and active neutron measurement data are processed through a mathematical imaging algorithm. The purpose of the imaging algorithm is to reduce the measurement uncertainty due to source positioning variation within the waste containers. The result is improved sensitivity and regulatory compliant precision and accuracy of results. [4].

The IPAN™ neutron measurement technique (figure 3) combined with GEA measurements and AK data have been successfully developed into analytical methodologies to perform WIPP certified measurements on containers of Contact Handled (CH) waste designated for disposal at the WIPP. These systems have been demonstrated to meet the sensitivity requirement to perform Low Level Waste (LLW)/TRU sorting capability at the 100 nCi/g level for both 55 gallon drums and for large waste crates [4].

Fig. 3 IPAN™ system

The first of these systems employed was the Mobile IPAN™ System, which has been performing WIPP certified assays and LLW/TRU sorting of 210 liter waste containers of CH-TRU on a routine production basis at the Savannah River Site (SRS) through contract with the Central Characterization Project (CCP) since 2001. This system employs the PAN/GEA/AK technique through the performance of an Expert Analysis associated with each assay.

The technique was further refined to expand the role of automated software to perform the combining of the PAN data with the GEA data on all but a small minority of "exception containers." This technique was implemented in the Multi-Purpose Crate Counter (MPCC). This System was developed to perform assays on Weapons-Grade plutonium waste contained in large waste crates at the Rocky Flats Environmental Technology Site (RFETS). The System has been performing production WIPP certified assays and LLW/TRU sorting of waste in these containers since 2002.

The Mobile IPAN™ System at SRS and the MPCC at RFETS have passed several WIPP audits and have complied with the WIPP Performance Demonstration Plan (PDP). Both systems have been designed for and continue to demonstrate compliance with the rigorous WIPP Quality Assurance Objectives (QAO's) for CH-TRU.

THE CASE FOR PAN/GEA/AK TECHNIQUE TO CHARACTERIZE RH-TRU

The characterization of RH-TRU for disposal at the WIPP will be conducted in compliance with Waste Acceptance Criteria, which at the present time have not been promulgated. It seems reasonable and prudent, however, to anticipate that such criteria will have the same levels of rigorous requirements as those specified for CH-TRU. Those requirements would include similar precision and accuracy objectives along with demonstrable and defensible determination of a lower limit of detection below the 100 nCi/g sorting level for LLW/TRU, participation in a demonstration program similar to the PDP for CH-TRU and rather rigorous requirements to verify isotopic AK.

The specific nature of RH-TRU present especially difficult challenges for any proposed technique for Waste Characterization. Some of these challenges merit discussion:

Many of the reportable WIPP isotopes have characteristic isotopes that can be measured by GEA with low detection limits in CH-TRU. The RH-TRU can severely alter these detection limits due to 3 effects: (i) The increased Compton continuum from high-energy gamma lines such as Cs137 and Co60, (ii) detector saturation from high total gamma activity waste may require introduction of an absorber (or filter), which further reduces the signal for low energy photo peaks, (iii) interfering isotopes such as Am243 create problems with photo peaks of interest with approximately the same energy.

High density and high "Z" matrices result in severe attenuation of gamma rays from low energy photo peaks of interest. This effect further limits the capability of gamma spectral analysis to quantify many of the peaks of interest.

The presence of non-gamma emitting nuclides such as Cm244 precludes the determination of these nuclides by gamma spectral analysis. Although Cm244 with an 18-year half-life is not classified as TRU, the isotope may dominate the passive neutron emission and contribute up to 95% of the radioactive hazard and thus must be characterized for compliance with transportation requirements.

While it could be seen as desirable from a cost and time consideration to adopt the most simplistic of NDA approaches to the characterization of RH-TRU, the difficult challenges addressed above become insurmountable challenges for the more simplistic approaches. If, for example a "gamma only" technique were considered, the problem of insensitivity to low energy photo peaks becomes an insurmountable challenge. The severe effect of the Cs137 and Co60 spectra on the LLD for the low energy photo peaks would raise the sensitivity to levels well above the required 100 nCi/g. The attenuation of the gamma lines from high density and high "Z" matrices would also have the same deleterious affects on the LLD. Without any other techniques, the "gamma only" approach would not be able to characterize gamma-silent nuclides.

The challenges of RH-TRU require special consideration in the PAN/GEA/AK technique, but the technique has a much greater flexibility and capability to meet these challenges than a more simplistic method. A discussion of that flexibility and capability in relationship to CH-TRU waste is merited.

For PAN systems, the capability to detect low levels of TRU (<100 nCi/g) in the presence of high levels of Cs137 and/or Co60 is only compromised if the high level of gamma radiation adversely affects the performance of the fast neutron detectors. It has been demonstrated that adverse affects of gamma radiation on these detectors can be compensated for by lead shielding to provide for essentially unaffected sensitivity at the radiation levels expected in CH-TRU [6]. Therefore, the capability to detect fissile isotopes in the range below 100 nCi/g can be maintained in CH-TRU without regard for the radiation levels from fission products in the waste stream.

The PAN/GEA/AK technique normally relies on the gamma analysis to provide isotopic fraction data for the majority of the TRU isotopes along with AK and mathematical correlation techniques to determine isotopes such as Pu242 in those waste containers with >100 nCi/g. That technique will be severely limited in cases where the Cs137 and/or Co60 activity precludes the detection of plutonium, uranium and americium isotopes. The same problem also exists when the matrix density attenuates the gamma spectrum sufficiently to render these isotopes undetectable. The methodology for mitigation of this problem becomes very closely linked with the specific waste streams, the specific regulatory requirements and the quality and nature of the AK associated with the waste. It does appear likely that for specific waste streams where the range of fuel burnup and uranium enrichment is not overly large that the isotopic composition (mass fractions) of the waste may be estimated using a combination of fuel irradiation histories and the computer inventory code ORIGEN. The utilization of these estimates will introduce additional uncertainty into the total measurement uncertainty (TMU). This must be evaluated for each applicable waste stream. If the estimates can be used to bound the TRU mass determination without exceeding the PDP quality objectives, the method should be defensible to the regulating authority. This argument would require specific justification and demonstration on an individual waste stream basis. If these estimates are not acceptable, the mass fraction data may need to be supplemented by sampling and destructive analysis techniques. This technique introduces additional sources of error, such as sampling error. These must be considered and the technique requires special approval by the regulating agency.

The presence of the non-gamma emitter Cm244 can be addressed in the PAN/GEA/AK technique by a comparison of the active neutron results and the passive neutron results. Since the Cm244 produces spontaneous neutrons, its presence is only seen in the passive result. If the isotopic fractions of all species contributing to the active and the passive neutron response are known values except for the Cm244 fraction, that value can be mathematically derived from the measured data.

The CH-WAC requires that Cs137 and Sr90 be determined and reported. The current PAN/GEA/AK technique utilizes mass fraction data to characterize Cs137 from the 662 keV photo peak. The Sr90 mass is bounded by the measured Cs137 and reported as such. In the case of potential RH-TRU, it is likely that the plutonium isotopes will not be detected and that the Cs137 mass fraction will not be known. For these circumstances, it is anticipated that the GEA will be expanded to provide for direct quantification of the Cs137 from the gamma spectral data using techniques already employed by "gamma only" analytical methods.

A tabular comparison of the capabilities of the PAN/GEA/AK technique and a Gamma Only NDA technique to address the challenges presented by CH-TRU are presented in Table I. In summary, the challenges presented by RH-TRU require special consideration in light of the specific waste streams and the regulatory requirements. Due to the flexibility and capabilities of the PAN/GEA/AK technique, it appears to be a much more feasible option to be tailored to handle RH-TRU waste characterization than more simplistic NDA techniques.

Table I. Comparison of PAN/GEA/AK Capabilities to Address Challenges Presented by RH-TRU.

RH-TRU Problem/Challenge	**PAN/GEA/AK Technique Solution to Problem**	**Gamma Only Solution to Problem**
Difficult to extract useful data from gamma spectrum due to: (i) High Compton Continuum from Fission Products precluding detection of low energy photo peaks and/or, (ii) Attenuation of Gamma peaks in high density and high “Z” matrices.	Utilize Active Neutron Data to Detect Fissile Isotopes at or below 100 nCi/g Utilize AK, fuel irradiation histories and ORIGEN code to provide isotopic fraction data. Last Resort: Develop scheme for sampling and DA to supplement AK mass fraction data. Active Neutron Data to detect Fissile Isotopes at or below 100 nCi/g.	Utilize sampling and DA techniques to ratio TRU Isotopes to Cs137 (the quantifiable parameter). Problematic due to inconsistency of Cs137 retention in the fuel processing may lead to high TMU.
Presence of Cm244	Compare Active Neutron Data and Passive Neutron Data and the mass fraction of all other active/passive neutron source isotopes to derive Cm244.	No gamma emission
Quantification of Cs137	Direct determination from the Gamma Spectrum.	Direct determination from the Gamma Spectrum

CONCLUSIONS

The unique combinations of circumstances created by specific TRU waste streams, their relationship to the body of associated AK, the range of containers and matrices in which they are contained and the regulatory requirements for their disposal create unique combinations of NDA requirements and challenges. These requirements and challenges have historically been addressed by tailoring an NDA technique to the unique circumstances created by the application.

The range of possible conditions for a specific waste stream may be so vast as to preclude complete automation of the NDA technique through software rule-based decisions and mathematical formulae. As seen in the historical applications of the PAN/GEA/AK technique, Expert Analysis may supplement the automated software. The Expert Analysis is initiated when the software flags an exception condition.

Each combination of waste stream, container-type and regulatory requirements create special considerations for the determination of TMU. The TMU must be included in the final reporting of characterization data. Therefore, the software must be tailored to address the specific TMU analysis for the waste stream and container.

Specific regulatory requirements or waste stream conditions may require that the physical configuration of the NDA equipment be tailored to a set of conditions. As examples, the chamber size may require special consideration for the waste container size or the number of detectors may be increased to increase sensitivity. When the physical configuration is tailored, the software must be tailored to accommodate the configuration.

As the regulatory requirements change or the scope of the application changes, the methodology may need to undergo additional tailoring to accommodate the changes.

Special or unique circumstances that have been encountered in specific waste streams that require tailoring of the System have included the presence of PuF_4 resulting in additional neutrons from alpha-n reactions, presence of nuclides not anticipated in AK, presence of heat-source plutonium (elevated Pu238 mass fraction), and the presence of neutron emitters including Cm244, Cm246 and/or Cf252. The analytical system in conjunction with Expert Analysis has provided sufficient flexibility and capability to achieve defensible estimates of TRU isotopes in all of these circumstances. Although RH-TRU fuel provides additional unique circumstances and challenges it is anticipated that this technique stands poised to meet these challenges through additional tailoring of the technique. The PAN/GEA/AK technique is, therefore, rather uniquely poised to meet the specific challenges of these waste streams and to satisfy the anticipated regulatory requirements for disposal of RH TRU.

REFERENCES

1. A. P. Simpson, "Meaningful Characterisation of High Beta Gamma Radwaste by Radiometric Instrumentation," International Conference on Radioactive Waste Management and Environmental Remediation, 1997.
2. P. A. Clark, et al, "Development and Application of Special Instrumentation for Materials Accountancy and Process Control in Spent Fuel Recycle Plants, INMM, Japan, 1996.
3. A. S. Chesterman, et al, "Spent Fuel and Residue Measurement Instrumentation at the Sellafield Nuclear Fuel Reprocessing Facility," INMM, 1995.
4. J. T Caldwell, et al, "High Sensitivity Imaging Passive and Active Neutron (IPAN) Transuranic Assay System," 14^{th} Annual ESARDA Meeting, Salamancia, Spain, May 1992.
5. M. J. Clapham, A. P. Simpson, et al, "Operation and Regulatory Performance of Waste Crate Assay Systems at RFETS," Waste Management, 2003.
6. D. H. Beddingfield, H. O. Menlove, et al, "He3 Neutron Proportional Counter Performance in High Gamma Ray Dose Environments," Nuc. Inst. And Methods in Phys. Research, A 455 (2000) 670-682.