SANDIA REPORT

SAND2005-6223
Unlimited Release
Printed October, 2005

Advanced Instrumentation for Reprocessing

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Prepared by
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Advanced Instrumentation for Reprocessing

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Abstract

Recent interest in reprocessing nuclear fuel in the U.S. has led to advanced separations processes that employ continuous processing and multiple extraction steps. These advanced plants will need to be designed with state-of-the-art instrumentation for materials accountancy and control. This research examines the current and upcoming instrumentation for nuclear materials accountancy for those most suited to the reprocessing environment. Though this topic has received attention time and again in the past, new technologies and changing world conditions require a renewed look and this subject. The needs for the advanced UREX+ separations concept are first identified, and then a literature review of current and upcoming measuring techniques is presented. The report concludes with a preliminary list of recommended instruments and measurement locations.

Acknowledgement

The author would like to thank Josh Phillips and Gary Rochau for their help in the research. Additionally Mark Schanfein, Ed Arthur, Tracia Love, and Paul Rexroth provided valuable feedback.

Acronyms

Elements

Am	Americium
Cm	Curium
Cs	Cesium
I	Iodine
Np	Neptunium
Pu	Plutonium
Sr	Strontium
Tc	Technetium
U	Uranium

Instrumentation

AES	Atomic Emission Spectroscopy
AXRF	Accelerator Stimulated X-Ray Fluorescence
CPC	Constant Potential Coulometry
DNL	Delayed Neutron Lifetime
HKED	Hybrid K-Edge Densitometry
HRGS	High Resolution Gamma Spectoscopy
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IDGS	Isotope Dilution Gamma Spectroscopy
IDMS	Isotope Dilution Mass Spectrometry
KED	K-Edge Densitometry
LED	L-Edge Densitometry
MS	Mass Spectrometry
NCC	Neutron Coincidence Counting
NICS	Neutron Integrated Cross-Section Spectroscopy
NMC	Neutron Multiplicity Counting
NRF	Nuclear Resonance Fluorescence
SGS	Segmented Gamma Scanning
TARIS	Thermal Atomization Resonance Ionization Spectroscopy
TIMS	Thermal Ionization Mass Spectrometry
XRF	X-Ray Fluorescence

Other

HPGe	High Purity Germanium
MOX	Mixed Oxide Fuel

Advanced Instrumentation for Reprocessing

Introduction

A renewed interest to reprocess spent nuclear fuel in the U.S. has led to a need to investigate materials accountability instrumentation. Reprocessing plants contain large inventories and a large variety of nuclear materials which must be managed and tracked following national and international guidelines. The current era of proliferation concerns associated with special nuclear materials and terrorism threats makes materials protection, accountability, and control more important than ever. This study investigates instrumentation in current reprocessing plants as well as advanced technologies that may be considered for new plants.

Background

There are currently a number of reprocessing plants around the world of various sizes. The largest are La Hague in France, Sellafield in Great Britain, and Rokkasho in Japan. All of these large-scale commercial facilities use the PUREX separations process to remove U and Pu from spent nuclear fuel. The U and Pu are recycled back into the fuel cycle to be reused in reactors. The leftover actinides and fission products are placed into a waste form for geologic burial.

The U.S. is investigating the advanced UREX+ processing concept which is similar to PUREX. UREX+ uses multiple separation steps to eliminate the major contributors of radioactivity and heat load from the high level waste. The major purpose of the multiple separation steps is to drastically decrease the heat load of waste destined for the Yucca Mountain repository both for increased safety and to significantly extend the repository capacity. Both Pu and Np are removed together to produce fuels for thermal reactors. This approach can offer some material tracking advantages for fresh fuel. Upon irradiation, a build-up of ^{238}Pu increases the intrinsic proliferation resistance of the used fuel.

Traditionally the quantities of U and Pu have been the main elements of interest for materials accountability since these products are separated. The UREX+ concept is going to require accounting of the additional separated elements, though the required level of precision is uncertain at this point.

Materials of Interest

An overall process flow diagram for the UREX+3 concept with full separations capability is shown in Figure 1. There are four major product streams: U, Pu/Np, Am/Cm, and Cs/Sr. The U and Pu/Np can be recycled back into light water reactors. The Am/Cm can be burned in fast reactors or using any high flux source of fast neutrons. The Cs/Sr can be placed in temporary storage for natural decay due to their 30 year half-lives. Finally, there may be some other isotopes like I, Tc, and tritium that should be accounted for due to their radiotoxicity and long-term dose characteristics.

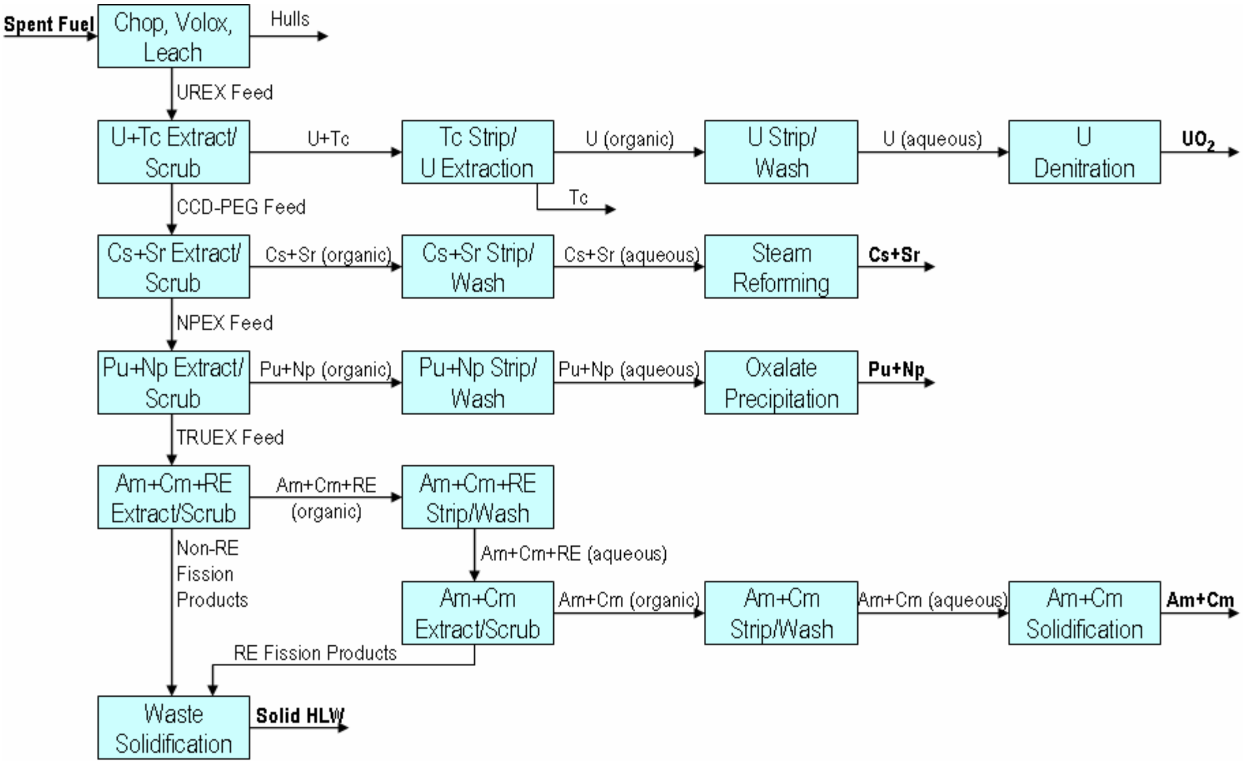

Figure 1: UREX+3 Flow Diagram

These materials must be measured with varying levels of accuracy upon receipt, while in storage or processing, and prior to shipment. In any process, accurate determination of the spent fuel composition is a difficult task due to the high radiation and heat levels, as well as the complexity of composition. The final product streams are easier to measure since the bulk of the radiation field is removed. The difficulty is in obtaining high precision at the front end.

Near Real-Time Accountability (NRTA)

Near Real-Time Accountability (NRTA) is a term used frequently in materials accountancy research. Current reprocessing plants do keep track of bulk quantities of material in near real-time. Standard industrial measures like level indicators on tanks and flow meters are used to keep track of total solution volume and throughput. However, the actual U and Pu content is measured by taking a sample to a laboratory for analysis, and this analysis is time-consuming. One of the goals of an advanced reprocessing plant is to accomplish NRTA of elemental and possibly even isotopic compositions in addition to keeping track of bulk quantities.

To determine a total inventory of a particular element, two measurements (or more) are often required. For example, to determine the total U quantity in a tank, a level indicator is used for volume, and a laboratory analysis is used to determine concentration. A level indicator easily takes NRTA measurements, but the concentration measure would have to be modified for on-line capability. Therefore, part of the long-term goal of this research is to investigate which of the precise laboratory instrumentation could be modified for automated sampling and measurement.

When two or more measurements are used to determine a quantity of interest, proper uncertainty analysis must be followed to determine the cumulative effects of all the measurement uncertainties. The smallest uncertainty of one measurement may place a limit on the feasibility of reducing another measurement down to very low uncertainties. Improving the uncertainty of the quantity of interest will require improving the accuracy of all instrumentation used to arrive at that quantity.

Current Instrumentation

The following presents a literature review of the various measurement techniques used in reprocessing. An overview of the technique is presented along with the usefulness, precision, sample type, and preparation required if the information was given. It is important to note that precisions listed may be specific to a certain experimental technique, so the numbers will be used to get a general idea of the value of the measurement. One difficulty with this review was that information was given as uncertainty, accuracy, precision, or variation depending on the author. All measurement systems have two error types: random and systematic. This review focused on the random error. An attempt was made to present all data in terms of the random error uncertainty, and it will be assumed that the uncertainty is that for one standard deviation.

Standard Industrial Measures

Level & Density Indicators

Level and density of a tank solution are measured by differential pressure measurements. Two probes are submerged in the solution at a separated distance, and a third probe is located at the top for the reference pressure. The difference in pressure between the two submerged probes determines the density of the fluid. The difference in pressure between the reference probe and the lowest submerged probe determines the level (and volume) of the solution.

Usefulness: Solution level, density, and volume measurements. Differential digital quartz pressure transducers have an uncertainty of 0.2%.[1]
Sample Type: In tank measure.
Prep: Adequate mixing required.

Flowmeters

Flowmeters could play an important role in materials accountability when the processing is continuous, but they have not been used for materials accountancy in the past. The current state-of-the-art in flow measurements will need to be investigated in more depth in future work.

Usefulness: All types of fluid flow. Standard industrial flowmeters have uncertainties ranging from 2-5% depending on piping dimensions. High precision flowmeters can reach 0.1% uncertainty under optimal design conditions.
Sample Type: On-line measure.
Prep: Long, straight sections of piping increase accuracy.

Nuclear Measures

K-Edge Densitometry (KED)

In KED, an x-ray spectrum is sent through a sample with a scintillation detector on the other side. K-shell electrons are liberated by absorbing x-rays of high-enough energy, and this absorption shows up as a drop in the relevant portion of the x-ray spectrum. The energy of the drop-off determines the particular heavy metal present, and the magnitude of the drop determines the concentration. The U k-edge is at 115.6 keV.[2] A similar technique, LED, measures the liberation of L-shell electrons.

Usefulness: U elemental concentrations, 0.2% uncertainty for 1000 s count (at 180 g/L).[3]
Sample Type: Dissolver solution, high U content solutions.
Prep: No chemical preparation required.

X-Ray Fluorescence (XRF)

An x-ray source (or other excitation source) is used to excite electrons. Each element has a unique spectrum of x-rays given off after excitation that can be used to determine the ratio of elements present. Alone, XRF can be used to determine Pu concentration to 2% uncertainty.[4] This method is usually used in conjunction with KED or another measurement technique that gives the concentration of either U or Pu to decrease the uncertainty (see next). A higher energy x-ray source may be able to reduce the uncertainty of the measurement by liberating higher energy electrons. This high-energy variation is called Accelerator Stimulated X-Ray Fluorescence (AXRF), and may be possible with the use of compact electron accelerators.

Hybrid K-Edge/X-Ray Fluorescence Densitometry (HKED)

HKED combines the previous two measurement techniques to give more accurate measures of Pu concentration. The same x-ray source is used for both measures. First, a KED measurement using a high purity germanium (HPGe) detector determines the U concentration. Then, a second HPGe detector measures the intensity ratio of $K\alpha_1$ x-rays from both U and Pu. The ratio is used along with the U concentration measurement to determine the Pu concentration.

Usefulness: U elemental concentrations, 0.2% uncertainty for 1000 s count (at 180 g/L);[3] Pu elemental concentrations, 0.7% uncertainty for 1000 s count,[2]
Sample Type: Dissolver solution, any solutions with actinides.
Prep: No chemical preparation required.

High Resolution Gamma Spectroscopy (HRGS)

HRGS is used to determine the gamma emission rates of nuclear material. Due to the large number of radioactive species in spent fuel, this measurement technique typically has lower precision when more species are present. It is more useful after species have been separated out from the fission products or for measuring low level waste. Segmented gamma scanning (SGS) is a method for using HRGS to measure non-homogeneous waste canisters.

Usefulness: 0.5-2% uncertainty for separated materials.[5]
Sample Type: Any separated stream, or low-level waste streams.
Prep: None.

Isotope Dilution Gamma Spectrometry (IDGS)

In IDGS, a diluted dissolver solution sample is measured using a high purity germanium detector along with a known Pu spike (sample) to determine Pu isotopic concentrations.

Usefulness: ^{240}Pu & ^{239}Pu concentrations, 0.2% uncertainty for ^{239}Pu concentration, 0.5% uncertainty for ^{240}Pu/^{239}Pu ratio.[6]
Sample Type: Dissolver solution, any other Pu solution.
Prep: Dilution of sample required, Pu spike needed for comparison.

Mass Spectrometry (MS)

MS is the traditionally accepted way to verify measurements with high accuracy. However, measurement time can be lengthy since the sample must be chemically separated before being placed in the mass spectrometer to eliminate isobaric interferences (^{238}Pu looks the same as ^{238}U). There are a number of different methods which have developed over the years: Isotope Dilution (IDMS), Thermal Ionization (TIMS), and Inductively Coupled Plasma (ICP-MS). ICP-MS has replaced the others due increased precision, and shorter preparation and analysis time. ICP-MS uses an argon plasma to ionize a sample. The ionized sample is then accelerated into a mass spectrometer. Current research continues to improve precision.

Usefulness: (IDMS) Pu concentration, 0.3-0.5% uncertainty.[3]
(TIMS) U and Pu isotopic ratios, 0.1% uncertainty.[5]
(ICP-MS) U and Pu isotopic rations, <0.1% uncertainty.[7]
Sample Type: Any solution.
Prep: Chemical separation required.

Passive Neutron Coincidence/Multiplicity Counting (NCC) and HRGS

NCC/HRGS uses two measurement techniques to determine Pu quantities. A NCC (or NMC) measures the spontaneous fission rate from the sample to determine the effective ^{240}Pu mass. HRGS is used to measure the isotopic ratio of ^{240}Pu to ^{239}Pu and other major Pu isotopes to determine total Pu mass.

Usefulness: Pu quantities, 2-5% uncertainty (mostly due to geometry and attenuation uncertainties).[3]
Sample Type: Dissolver solutions, Pu content in glove boxes, holdup, product, MOX canisters, and waste.
Prep: None.

In addition to passive neutron counting, active neutron counting uses an external neutron source to initiate fission events. The resulting neutron release and decay can be measured. Techniques include Neutron Integrated Cross-Section Spectroscopy (NICS) and Delayed Neutron Lifetime (DNL). These techniques will need to be investigated further, but active neutron counting typically has lower precision, and will probably not be applicable to advanced accountability.

Constant Potential Coulometry (CPC)

CPC is used to determine total Pu in product solutions. In this technique, an amount of electricity is passed through a solution and measured. The resulting measurement is directly related to the analyte concentration.

Usefulness: Total Pu, 0.1-0.2% uncertainty.[3,5]
Sample Type: Pu product solution.
Prep: Requires laboratory preparation, interfering chemicals must be removed.

Titrimetry

Titrimetry is used to determine Pu or U in product solutions. The volume of a solution of known concentration is used to determine the quantity of analyte.

Usefulness: Total Pu, 0.1-0.5% uncertainty.[3,5]
Total U, <0.1% uncertainty.[5]
Sample Type: Pu or U product solution.
Prep: Requires laboratory preparation.

Gravimetry

Gravimetry is the quantitative measurement of an analyte by weighing a pure, solid form of the analyte. Obtaining pure solids from solutions containing an unknown amount of a metal ion is done by precipitation. The total weight along with the initial volume determines the density.

Usefulness: Total Pu or U, <0.1% uncertainty.[5]
Sample Type: Pu or U Oxide.
Prep: Requires laboratory preparation.

Spectrophotometry

Monochromatic light is passed through a sample, and the decrease in intensity on the other side determines the absorption of the light in that sample. Each compound has a unique absorption wavelength.

Usefulness: Pu or U content, 0.2-0.3% uncertainty for product solutions, 1.0% for dissolver solutions.[3]
Sample Type: Pu or U product.
Prep: None.

Atomic Emission Spectroscopy (AES)

In AES, a sample is atomized within a vacuum chamber. The atoms are excited, and the light emitted is used to determine the element present. A high-resolution spectrometer can distinguish the wavelengths from one another. This technique is not as useful for analysis in the input and dissolver solutions due to the dominance of the U isotopes. After U extraction, though, it may be more useful for identification of a wide variety of elements. More investigation of this technique is required.

Calorimetry

Calorimetry is useful for Pu/Am assay of products and wastes by measuring the heat output. The measurement is non-destructive and accurate. Accurate isotopic ratio data is needed to determine the quantities of interest, so for accurate measurements mass spectrometry is required.

Usefulness: Pu or Am content, ~0.1-0.5% uncertainty depending on product.[9]
Sample Type: Pu or Am product.
Prep: None.

Chromatography

Chromatography is used to determine the concentration of U in mixtures. Liquid chromatography is suitable for process control, but not for materials accountancy, so will not be examined in detail.

Plutonium Alpha Monitors

Alpha monitors can be used to determine rough amounts of Pu. These monitors are usually used for criticality control or for indications of Pu in waste streams. The uncertainty is usually in the range of 5-10%, so these monitors are not useful for materials accountancy.

New Instrumentation

Thermal Atomization Resonance Ionization Spectroscopy (TARIS)

TARIS is a more automated mass spectrometry system that eliminates the chemical preparation usually required with mass spectrometry. TARIS has been developed by Atom Sciences, Inc.[10] and combines three technologies. The input solution is first atomized on a hot tantalum plate into a vacuum chamber (Thermal Atomization). Then a laser is used to selectively ionize a particular element group (Resonance Ionization). Finally, that element group is sent into a mass spectrometer to determine isotopic quantities.

The potential advantage of TARIS is the ability to achieve the precision of mass spectrometry in an automated system that can be designed to measure an isotopic spectrum for multiple element groups. Because the detection does not use gamma rays, the large radiation spectrum in dissolved fuel does not interfere with the measurements. The major disadvantage is cost, but for a large reprocessing plant, the added precision may be worth the cost.

The resonance ionization step is fairly involved. For each element, two laser wavelengths are needed to first excite and then ionize the excited atoms. This means two lasers are needed for each elemental spectrum of interest. If multiple elemental analyses are desired, a tunable laser may make more sense. The cost of TARIS was quoted as $1 million for a unit capable of measuring one element, and an additional $0.5 million for each additional set of lasers for additional elements.[10] The tunable laser may be the better choice if it is desired to measure the isotopic spectrum of U, Pu, Np, Am, Cm, Cs, and Sr.

The overall detection efficiency of TARIS is about 3-5%, but this should not be a problem for input solutions that contain significant quantities of materials. Precision is estimated to be in the range of <0.1%. The device is believed to be reliable allowing for measurements taken every 10-20 minutes. Maintenance will be required at least every 6 months.

Thermal atomization is suited for liquid samples, but it is also possible to use an ion beam to sputter a solid material. It may be worth examining the use of TARIS to measure solid spent fuel, but likely the non-uniformity of the fuel pieces will make this technique unreliable. TARIS seems suited to use whenever the isotopics of a liquid stream are desired.

Atom Sciences is interested in coordinating further development of this technology. TARIS would probably be most useful measuring the dissolved fuel at the front end of the reprocessing plant (since other techniques have problems at this location). A first step may be to assist with testing at Atom Sciences using non-radioactive samples. For example, creating a dissolved solution of heavy and other metals that contains stable species could be used to determine the precision of TARIS and to make sure no interferences occur.

The next step that would be desired is laboratory testing using more representative samples. If dissolved nuclear fuel is used, this research will have to be performed at a national laboratory within a hot cell. This testing will require a significant investment due to the cost of the equipment and cost for running such an experiment.

The final step will be to install and test the instrument on the Advanced Fuel Cycle Facility. Before this point, a decision will need to be made as to which elements the system should be able to measure. Standard equipment will be used on the Advanced Fuel Cycle Facility for materials accountability, but the facility will be designed with places to test advanced equipment like TARIS.

Nuclear Resonance Fluorescence (NRF)

NRF uses a high energy photon spectrum (2-8 MeV) to excite nuclear states which are unique to every isotope.[11] The resulting de-excitation of the nucleus releases a high energy photon that can be detected. Unlike atomic excitation, nuclear excitation occurs with much fewer wavelengths to decrease interferences. This technique may be a non-intrusive way to determine isotopic abundances in reprocessing samples.

NRF requires a high energy photon source which can be created using a compact electron accelerator. The cost of this unit may be comparable to TARIS, but only one accelerator and detector are required for multiple elements of interest. An advantage over TARIS is that it could be used for solid samples (for the incoming fuel assemblies). The penetrating power of the high energy photons allows for a non-uniform sample to be measured. This solids measurement may be the most promising application of NRF.

There are a few uncertainties with this measurement technique. First, the nuclear resonance states are not completely known for all isotopes of interest. Lengthy experimental work could be required to build up a database. Second, since the measurement technique is gamma detection, there may be interferences from the background radiation spectrum. Finally, there is no data on the precision of this type of instrumentation. The originally suggested use is for detection of nuclear materials in cargo containers, in which case specific quantities are not necessary.

Due to the previous concerns, there is significant development work required for this technology. The first step is to determine what nuclear data is needed for the reprocessing application, and how that data can be obtained experimentally.

Preliminary testing could begin at the Idaho Accelerator Center which has compact electron accelerators for use in creating the desired photon spectrum. Benign, non-radioactive species for which nuclear resonance data is available can be tested first. Of particular interest is determining the precision of the instrumentation. A sample containing multiple metals and heavy metals would be useful to test to look for any possible interferences.

Should the technique prove promising, the next step would be to test radioactive species. Again, hot testing will need to be performed at a national laboratory if a representative spent nuclear fuel sample is used. It will be important here to test the effect of the radiation spectrum on the results.

Quantum Dots

A much newer technology option for materials accountability is commonly referred to as quantum dots. Quantum dots are nano-scale crystals of semiconductors which bond to specific molecules and compounds. The dots emit a fluorescent light that can be tuned by changing the size of the particle by only a few nanometers. Therefore, the dots form a fluorescent tag specific to a particular element or molecule. This technology is most widely used in the biomedical field.

In the PUREX process quantum dots could be made to bond to U or Pu nitrate in order to identify the relative concentration of each. However, since this is such a new technology, there are many unknowns. The precision of the detection and analysis is not clear, especially if there are background effects that could interfere with the fluorescence signal. It is necessary to determine whether a small sample should be extracted for analysis, or if the dots could simply be added to the stream and monitored in real-time as the material passes through the various separation processes. The rinsing method to extract the dots after use would need to be developed. There are also concerns about the effect of radiation on the dots.

A small level of effort is underway at Sandia National Laboratories to investigate this technology, but too little is known about the durability of this technique to recommend it at this time. The research will be followed to determine if it could be useful for future assaying methods.

Measurement of UREX+ Streams

The increase in separation steps of UREX+ will require developing the available accountancy instrumentation to keep track of Np, Am, Cm, Cs, Sr, and possibly other species. The fissionable species accountancy is driven by safeguards, but the fission product accountancy will probably be driven by waste characterization needs. The following presents a first step to determining accountability of these materials.

^{137}Cs Determination

Cesium is a dominant heat source term in spent nuclear fuel due to its 30 year half-life. As such, measurement of this isotope can be used to determine the total heat output of fuel rods. In standard burnup, standard enrichment fuel near 20 years in age, ^{137}Cs can be clearly measured using high resolution gamma spectroscopy. ^{137}Cs goes through beta decay that results in the production of ^{137m}Ba, which has a 2.55 minute half-life. The 662 keV gamma released by the decay of ^{137m}Ba is used to quantify the amount of ^{137}Cs present. The uncertainty from this technique is near 1.4%.[12] This simple gamma measurement could be used to quantify Cs both in the dissolver solution (or spent fuel form) and in the final Cs/Sr product stream.

^{237}Np Determination

^{237}Np cannot be determined using HKED in the dissolver solution since the concentration is too low, and because the concentration of U is so high. It also cannot be measured by HRGS because the gamma ray released by ^{237}Np is about 3-5 orders of magnitude lower than the rest of the fission product spectrum. However, after the Pu/Np extraction step, the ^{237}Np concentration can be determined using HKED and HRGS. HKED is used to determine the concentration of Pu, and HRGS is used to determine the ratio of ^{237}Np/Pu—combining both gives the Np concentration.[13]

^{237}Np can be measured using the 86.477 keV gamma released by direct decay, or by measuring the 300 keV gamma released by decay of its daughter, ^{233}Pa (if equilibrium is reached). Using this technique, the ratio of Np/Pu can be determined with an uncertainty of 5%.[13] Note that Np may not need to be accounted for with as much precision as Pu and U due to lower mass throughput. Regardless, Np will need to be accounted for prior to Pu/Np extraction, as well as quickly verified afterwards and through holdup measurements. Current measurement techniques will need to be improved to effectively measure Np.

Am/Cm Determination

Americium and Curium can be measured using Isotope Dilution Gamma Spectrometry once the elements are separated from the waste stream. Before that point, gamma spectrometry is useless since the Am/Cm gamma rays are masked by other species. Measurement in the product stream may be able to reach an uncertainty of <0.9%.[14]

It is also possible to determine Am/Cm concentrations in the input solution by alpha-spectrometric methods. The alpha activity ratio of Am to Pu and Cm to Pu can be used along with the concentration of Pu from HKED to determine amounts of Am and Cm. Experimental work on this technique[15] used lengthy sample preparation steps and measured the isotopes alone without any other interferences. It may be difficult to apply this technique to the reprocessing environment with the full fission product spectrum.

Preliminary Conclusions

Based on the research done here, a preliminary list of instrumentation recommendations for UREX+3 was developed. This list is by no means complete, and it is more of a recommendation for further research rather than design choices for an advanced reprocessing plant. An overall observation is that it is much easier to measure elements or isotopes using established instrumentation once the streams are separated out. Therefore, the focus of new instrumentation is on the front end of the plant.

Most of the high-precision measuring techniques shown in the following list are currently laboratory measures. For near real-time accountability, these laboratory techniques need to be modified for on-line operation.

Spent Nuclear Fuel
HRGS can be used to verify burn-up calculations at a moderate level of precision.
NRF should be investigated as an alternative measurement.
Active and passive neutron techniques should be investigated.

Dissolved Fuel
HKED should be used for U/Pu concentrations, and other elements should be investigated.
TARIS & NRF should be investigated for determination of all elements/isotopes of interest.
ICP-MS should be used for concentration and isotopic abundances (laboratory analysis).

Intermediate Processes
HKED can apply in many locations.
IDGS or HRGS can be used for concentration and isotopic abundances.

U Product
Titrimetry, CPC, or KED should be used for total elemental amounts.
HRGS should be used for isotopic abundances

Pu/Np Product
Titrimetry, CPC, or KED should be used for total elemental amounts.
NCC/HRGS should be investigated for total Pu inventory.

Cs/Sr Product
HRGS can be used for cesium determination.
Sr assay needs more investigation.

Am/Cm Product
HKED should be investigated for concentration.
IDGS or Alpha Spectroscopy should be investigated for isotopic abundances.

Waste Streams
HRGS or IDGS should be used.
NCC/HRGS should be investigated along with the segmented gamma scanning.

Holdup Measurements
NCC/HRGS should be investigated for total Pu inventory.

Based on this preliminary research, three nuclear measuring techniques are suggested for further research. HKED is a non-destructive technique useful in many areas of the reprocessing plant. It would be advantageous to automate HKED to create an on-line technique for measuring elemental quantities. TARIS could be an extremely beneficial measuring technique at the front end to give precise measures of elemental and isotopic concentrations of many species of interest. The device will be costly, but the added materials accountability may be worth it. HRGS is used often, so standardizing the equipment could be beneficial due to its usefulness in many areas of the plant.

The standard measures of level, density, and flow meters should also be examined since these measures are used to determine inventories. More research is needed on NRF and Neutron Counting techniques to determine their usefulness for high-precision accountability. The recent advances in ICP-MS for verification should also be investigated. A number of techniques have been presented here, but it is probably not a good idea to have many different measuring techniques in the plant. Attention should be given to uniformity of equipment to help in the ease of correlating data, and to reduce costs.

References

1. T. Hosoma & Y. Maruishi, “Accurate Volume Measurement System for Plutonium Nitrate Solution,” *Journal of Nuclear Science and Technology,* v. 30, no. 5, p. 425-435 (May, 1993).
2. “Simultaneous Determination of Uranium and Plutonium in Dissolver Solutions from Reprocessing Plants — Combined Method using K-Absorption Edge and X-ray Fluorescence Spectrometry,” International Organization for Standardization, ISO 13464:1998(E) (April, 1998).
3. T.K. Li et al., “Evaluation and Development Plant of NRTA Measurement Methods for the Rokkasho Reprocessing Plant,” LA-UR-95-2276, INMM 36th Annual Meeting, (July 12-19, 1995).
4. H.A. Dayem et al., “Demonstration of Near-Real-Time Accounting: the AGNS 1980-81 Miniruns,” LA-9942 (January, 1984).
5. S. Deron & D. Donahue, “An Update of IAEA Analytical Capabilities for Safeguards Goals, Results and Challenges,” *Journal of Nuclear Materials Management*, v. 28, no. 2, p. 27-38 (2000).
6. T.K. Li et al., “Conceptual Designs of NDA Instruments for the NRTA System at the Rokkasho Reprocessing Plant,” LA-UR-96-2572, INMM 37th Meeting, (July 28-31, 1996).
7. J.S. Becker, “State-of-the-art and progress in precise and accurate isotope ratio measurements by ICP-MS and LA-ICP-MS,” *Journal of Analytical Atomic Spectrometry*, vol. 17, no. 9, p. 1172-1185 (September 2002).
8. S. Fiarman & R.B. Perry, “Attributes Measurements by Calorimetry in 15 to 30 Minutes,” *Nuclear Materials Management*, v. 16, p. 754-759 (1987).
9. “Standard Test Method for Nondestructive Assay of Plutonium, Tritium and ^{241}Am by Calorimatric Assay,” ASTM Standard C1458-00 (March, 2000).
10. T.J. Whitaker, Atom Sciences, Inc. (private communication), (August, 2005).
11. W. Bertozzi & R.J. Ledoux, “Nuclear Resonance Fluorescence in Non-Intrusive Cargo Inspection,” not yet published (2005).
12. P. Jansson et al., “Gamma-Ray Spectroscopy Measurements of Decay Heat in Spent Nuclear Fuel,” *Nuclear Science and Engineering,* v. 141, p. 129-139 (2002).
13. A. Morgenstern et al., “Analysis of ^{237}Np in Spent Fuel Solutions,” *Radiochimica Acta,* v. 90, p. 389-393 (2002).
14. W. Raab & F. Sus, “Determination of Microgram and Submicrogram Amounts of Americium in Spent Nuclear Fuel and Plutonium Samples by Isotope Dilution Gamma Spectrometry,” *Kernenergie*, v. 32, no. 2, p. 65-68 (February, 1989).
15. N. Shinohara & N. Kohno, “A Comparative Study of Alpha-Spectrometric Methods for Determination of Americium and Curium in Spent Fuel,” *Applied Radiation and Isotopes*, v. 40, no. 4, p. 346-348 (1989).

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