

Nondestructive Waste Assay Using Combined Thermal Epithermal Neutron Interrogation

Mixed Waste Focus Area



Prepared for
U.S. Department of Energy
Office of Environmental Management
Office of Science and Technology

December 1998

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OST Reference #1568

Mixed Waste Focus Area



Demonstrated at
Los Alamos National Laboratory
Los Alamos, New Mexico



Purpose of this document

Innovative Technology Summary Reports are designed to provide potential users with the information they need to quickly determine if a technology would apply to a particular environmental management problem. They are also designed for readers who may recommend that a technology be considered by prospective users.

Each report describes a technology, system, or process that has been developed and tested with funding from DOE's Office of Science and Technology (OST). A report presents the full range of problems that a technology, system, or process will address and its advantages to the DOE cleanup in terms of system performance, cost, and cleanup effectiveness. Most reports include comparisons to baseline technologies as well as other competing technologies. Information about commercial availability and technology readiness for implementation is also included. Innovative Technology Summary Reports are intended to provide summary information. References for more detailed information are provided in an appendix.

Efforts have been made to provide key data describing the performance, cost, and regulatory acceptance of the technology. If this information was not available at the time of publication, the omission is noted.

All published Innovative Technology Summary Reports are available on the OST Web site at <http://OST.em.doe.gov> under "Publications."

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SECTION 1

SUMMARY

Technology Summary

The Combined Thermal/Epithermal Neutron (CTEN) project was supported by the Mixed Waste Focus Area (MWFA) to develop and build a neutron based waste assay instrument that extends the capabilities of the baseline differential dieaway technique/passive active neutron (PAN) instruments, thus providing improved transuranic (TRU) waste assay accuracy.

The nondestructive waste assay capability needed to support Department of Energy (DOE) mixed waste characterization needs is necessarily a function of the waste form configurations in inventory. These waste form configurations exhibit a number of variables impacting assay system response that must be accounted for to ensure valid measurement data. Such variables include: matrix density, matrix elemental composition, matrix density distribution, radioactive material radionuclidic/isotopic composition, radioactive material physical/chemical form, and geometrical distribution in the waste matrix. The accuracy of TRU waste assay using the active DDT technique depends upon significant corrections to compensate for the effects of the matrix material in which the TRU waste is located. The CTEN has been designed to improve on PAN capabilities to better correct for the matrix and source effects on the measurement. Experimental results showed that for some matrices, corrections for position dependent effects within the matrix are possible. The enhanced capabilities that were designed into the CTEN system include:

- active and epithermal neutron interrogation for detection of fissile material self-shielding,
- new type of neutron multiplicity module for both active and passive measurements,
- detectors and methods to determine the distribution of fissile material in a waste drum (localization),
- Pulse-Arrival-Time Recording Modules (PATRM) for list-mode active and passive neutron coincidence counting,
- flux monitors to detect matrix inhomogeneities,
- methods to use the additional information to improve assay accuracy.

A summary of the CTEN instrument's demonstrated capabilities and the issues that will affect its implementation by a characterization facility is provided in this report to support end users and other interested parties in technology selection.

Demonstration Summary

Several types of testing were completed with the CTEN to calibrate the system and evaluate its performance. The basic data gathering tests used to complete the calibration and evaluation of the CTEN were:

- evaluation of four different active analysis methods: no correction, corrections with no positional correlations, corrections with positional correlations based on CTEN indicators, and corrections with forced positional correction based on Tomographic Gamma Scanner (TGS) images;
- determination of the accuracy of the CTEN lump correction method when applied to plutonium (Pu) or uranium (U) in a few representative drum-matrix types,
- evaluation of the passive multiplicity analysis to verify the sensitivity and accuracy of the passive assay for select matrix types,
- demonstration of CTEN on a selection of real waste drums from Los Alamos National Laboratory's (LANL) TA-55 to verify the absence of obvious unanticipated difficulties in applying the system to real waste.

All the tests described above concern CTEN methodology and involve straightforward CTEN assays on test samples. A variety of surrogate and actual waste drums and sources were used to perform these tests and are listed below.



Mock waste drums

Empty drum

Moderating drum (7.7 kg of hollow polyethylene balls, $\sigma_H = 0.0005 \text{ g/cm}^3$)

Iron drum (170 kg of iron scrap)

Ethafoam drum [Performance Demonstration Program (PDP) drum]

Combustibles drum (PDP drum)

Sources

3-mil highly enriched uranium (HEU) foils (mass range = 7-8 g ^{235}U each)

Low-burnup Pu disc (0.218 g ^{239}Pu)

3-g HEU sphere

10-g HEU sphere

100-g Pu cylinder

PDP distributed Pu sources

PDP lumpy Pu source

Twenty-five waste drums were subjected to CTEN active and passive assays with a lump correction for the active assay during the real waste demonstration. The waste matrices tested were the following:

10 – plastic/kimwipes

1 – rubber

1 – graphite

6 – nonactinide metal

1 – high-efficiency particulate air (HEPA)

2 – paper, wood, and rubber

2 – glass

1 – salt chloride

1 – salt chloride oxide.

Key Results

- A factor of 2.4 and 5.7 times fewer assay failures are seen at the 25 and 50% accuracy levels, respectively, with the CTEN spatial correction than with the best possible drum-averaged correction. In practical terms, this means that more difficult waste forms can be assayed using the CTEN passive mode than can be assayed using other passive counting methods, and that higher effective drum loadings can be certified for a given waste form.
- The CTEN spatial correction resulted in a factor of 6.8 times fewer assays falling outside the important 25% Waste Isolation Pilot Plant (WIPP) uncertainty limits than with the best possible drum averaged correction. No cases fell outside the 50% uncertainty limit versus 0.5% for the best possible drum-averaged correction.
- The lump correction is limited to use with HEU. This fills a gap in current nondestructive assay (NDA) capability since passive neutron counting gives good assays of lumpy weapons grade (WG) Pu, but cannot assay HEU. The 4helium (He) detectors used in the epithermal interrogation have a poor sensitivity, so at least 3 g of HEU (finely divided equivalent) are needed to perform reliable corrections.
- Tomographic Gamma Scanner (TGS) measurements were useful in determining how well CTEN's gross position indicators worked in locating radioactive material. The goal was not to necessarily pinpoint the contamination, but to determine whether material is located in the inner or outer radial regions, and top, middle, or bottom sections. By viewing TGS scans it was determined that this limited localization goal was indeed accomplished. The range of possible calibration factors was reduced for the CTEN resulting in a significant improvement in measurement accuracy.



Results of the real waste demonstration did allow for the following conclusions to be made:

- The real waste demonstration did identify a mislabeled drum. Although labeled as containing only Pu and previously assayed by passive neutron coincidence counter (PNCC) as containing 31.8 g of Pu, the active and passive CTEN assay results suggested that the drum might contain curium (Cm) rather than Pu. This was verified by examination of the cumulative gamma-ray spectrum of that drum from the TGS assay. In spite of the presence of the Cm, the CTEN was able to make a determination of the Pu content using the active mode (gamma assays can only measure to nominally about 500 mg).
- A significant trend identified from the real waste demonstration was that the simple spatial correction technique used in CTEN is approximately correct. This was validated by comparing the CTEN corrected data to the TGS assay results.
- One drum assayed was approximately 3/4th full with a more or less uniform matrix of NaCl, with a concentration of Pu near the upper surface of the matrix, where it is apparently only half shielded by the matrix. Although rare, cases where the matrix is nonbenign and heterogeneous, with a large fraction of the SNM positioned in such a way that the matrix interference is either much less or much more than is probable, can cause potentially large active assay positional errors. Once the matrix in the top 1/4th of the drum was forced to be air, the CTEN active assay value was improved.

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SECTION 2

TECHNOLOGY DESCRIPTION

This project consisted of both hardware and software development and optimization. A description of the hardware and software is given below.

Overall Process Definition

The widely-used active neutron DDT method uses thermal neutrons to “interrogate” fissile isotopes in waste drums. The resulting (induced) fast fission neutrons are detected in cadmium-shielded ^3He detectors that are insensitive to the interrogating thermal flux.

The CTEN method is similar to the DDT method, but interrogates the sample with both thermal and epithermal neutrons. This is achieved partly by the addition of ^4He detectors, which have a faster response than ^3He and can detect fast fission neutrons in the presence of the epithermal interrogating flux, and by a redesign of the moderating cavity so that thermalization occurs more slowly. Because epithermal neutrons are more penetrating in fissile material than thermal neutrons, the differential response can be analyzed to detect the occurrence of self-shielding by fissile material and measure the size of the effect. Self-shielding occurs when discrete lumps of fissile material are present, and can result in assay errors of several hundred percent. A fully operational CTEN device will perform all the functions of existing PAN devices with the added capability of being able to identify and assay lumps of fissile material. Additional features of the CTEN, not necessarily available in the current generation of PAN devices are the following: a neutron multiplicity measurement capability, increased active neutron detection efficiency, and a capability to detect nonuniform matrices and SNM distributions.

Technology Description

The CTEN instrument was designed and built at LANL in an effort to improve measurement accuracy and reduce the limitations of the DDT assay (see Figures 1 and 2). Modifications were made in the CTEN system’s hardware and software to improve active and passive measurement performance and the lump correction performance. The CTEN modifications include:

- List Mode Module – PATRM’s were developed to record the time of arrival of all neutrons, and the detector channel in which it was detected, in a list format (capacity of 1 to 4 million words). Thus, both timing and spatial information are recorded in one data collection path. The data can then be transferred to computer memory where it can be manipulated in any number of ways. For example, it can be scanned by “software shift register” or a “software multichannel scaler.” Other types of data manipulation and analysis, which can not be performed with conventional electronic modules are: the detection and rejection of cosmic-ray background neutrons, the determination of detector deadtime characteristics, and the detection of double pulsing and noise bursts.
- Epithermal Neutron Interrogation – To overcome self-shielding problems identified in DDT systems, the CTEN instrument observes the fissions that are induced by the epithermal neutrons that are present at earlier times after the neutron burst. The fission interaction probabilities for epithermal neutrons are typically an order of magnitude smaller than for thermal neutrons. Thus, the epithermal neutrons can penetrate further into lumps of fissile material, mitigating the effects of self-shielding.
- The CTEN instrument was designed to increase the length of time that epithermal neutrons are present. ^4He detectors were included in the design to count the fission neutrons during the epithermal interrogation.
- Passive Multiplicity Measurements - An advanced multiplicity analysis of the passive (^{242}Pu) neutron decay in Pu gives improved sensitivity and accuracy over the scaler and shift-register techniques used in existing PAN systems. Also, signal losses in the emitted spontaneous fission neutrons are automatically corrected for with this technique (the scaler and shift-register methods require matrix corrections as an additional step). The CTEN approach is similar to the “add-a-source” approach used in the High Efficiency Neutron Counter (HENC), but uses a passive matrix correction based on



active flux monitor and source-dependent positional parameters, rather than the transmission of a ^{252}Cf source.

- Active Multiplicity Measurements - The advanced multiplicity analysis performed on the active data, automatically corrects for signal losses for the emitted induced-fission neutrons, but requires a correction for attenuation of the interrogating neutron flux in the matrix and in lumps of SNM. The CTEN lump correction uses DDT-type active gated-scaler data, rather than the PATRM list-mode data used in the active multiplicity analysis; this gated-scaler data can be used to estimate masses in the same way as done in existing PAN systems.
- Neutron Imaging – The CTEN uses imaging to determine the spatial distribution of fissile material in a waste container. For a known matrix containing significant amounts of moderating or absorbing materials, the instrument response is dependent on the location of the fissile material in the drum. If the location can be determined, a position-dependent calibration factor can be applied to the observed response. To compare the CTEN to existing DDT instruments, an active measurement in a typical second generation DDT instrument obtains one data point for two time windows (early and late gate) for 14 detector packages for a total of 28 data points. The drum is rotated several times during the measurement to “average out” any angular nonuniformities in response. The CTEN instrument has 32 detector packages, five time windows, and data collected at 12 annular increments for a total of 1,920 data points for one active assay.
- Matrix Corrections - Separate DDT-type neutron interrogations are performed using epithermal and thermal neutron spectra (traditional DDT uses thermal interrogation only). Because the neutron absorption losses are less for epithermal interrogation than for thermal interrogation, the ratio of mass estimates for the two cases will vary with the drum matrix type and with the degree of self-shielding in lumps of the Pu or U. The epithermal/thermal mass ratio can then be used to estimate a matrix correction factor or if the matrix is known (or the system has been calibrated to that matrix type), the ratio is used for estimating lump corrections.

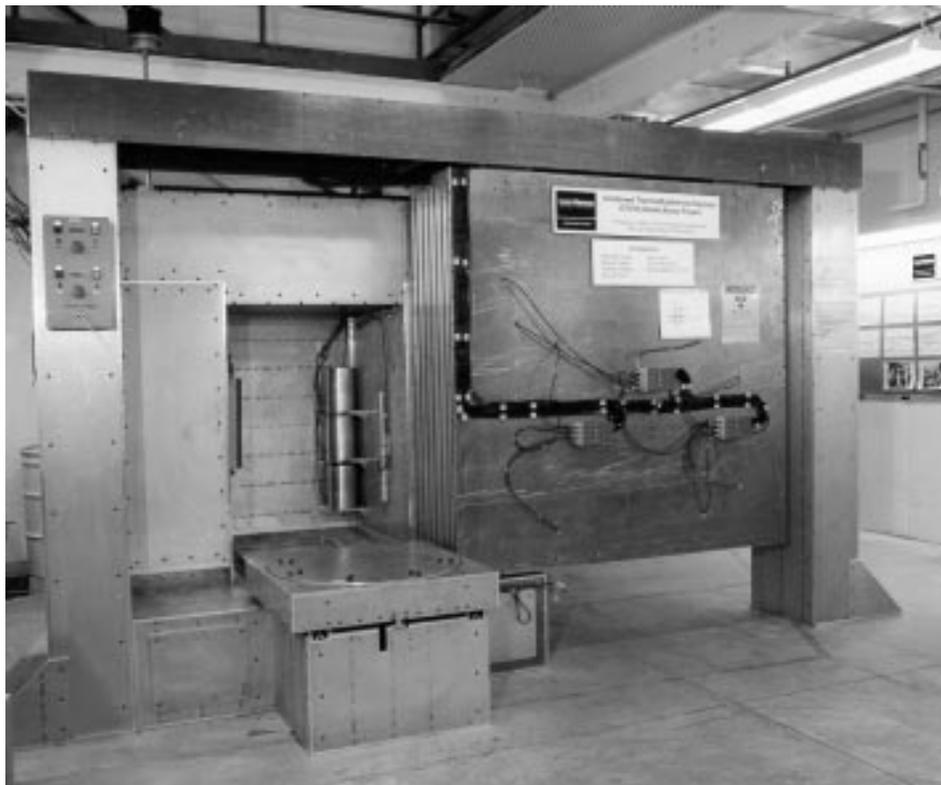


Figure 1. CTEN system.



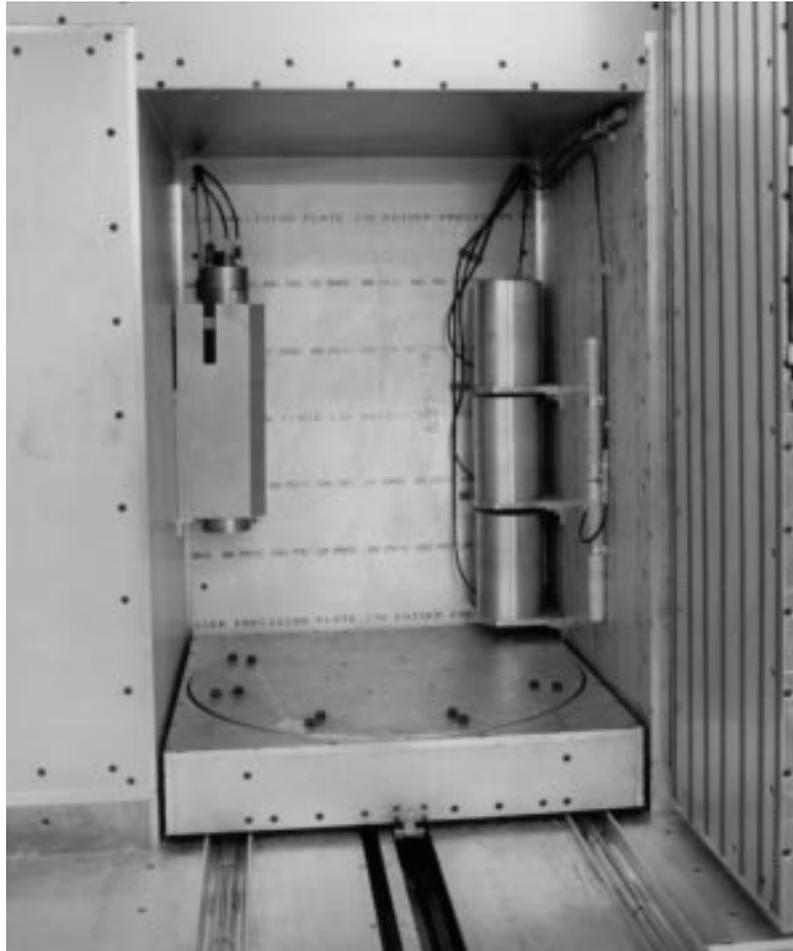


Figure 2. Assay chamber monitors.

SECTION 3

PERFORMANCE

Test Plan

Several types of testing were completed with the CTEN to calibrate the system and evaluate its performance (Estep and Melton September 1997; Estep et al. September 1998). The basic data gathering tests performed under this development effort are grouped into the following categories:

- active assay performance
- passive assay performance
- lump correction performance
- CTEN performance with real waste.

Table 1 summarizes the surrogates and sources used during the test phase to evaluate active, passive, and lump correction performance. The identifiers that were generated for the matrix drums and sources used in the various test cases and are listed below:

CTEN project mock waste drums

- A) empty drum,
- B) moderating drum (7.7 kg of hollow polyethylene balls, $\sigma_H = 0.0005 \text{ g/cm}^3$),
- C) iron drum (170 kg of iron scrap).

PDP drums

- D) ethafoam drum,
- E) combustibles drum.

CTEN project sources

- A) through D) 3-mil HEU foils (mass range = 7-8 g ^{235}U each),
- E) low-burnup Pu disc (0.218 g ^{239}Pu),
- F) 3-g HEU sphere,
- G) 10-g HEU sphere,
- H) 100-g Pu cylinder.

PDP sources

- I) - K) PDP distributed Pu sources,
- L) PDP lumpy Pu source.

Real waste drums

The waste matrices tested were the following:

- 10 – plastic/kimwipes
- 1 – rubber
- 1 – graphite
- 6 – nonactinide metal
- 1 – HEPA
- 2 – paper, wood, and rubber
- 2 – glass
- 1 – salt chloride
- 1 – salt chloride oxide.



Table 1. Mock-drum test cases for CTEN analysis (Note: The test categories overlap)

| Test purpose | Drums | Sources |
|------------------------------------|-------|---------|
| General lump correction | A – E | A – L |
| General passive analysis | A – E | H – L |
| General active analysis | A – E | H – L |
| Sensitivity | A – C | A, E |
| Lump correction, upper size limits | A – C | F, G, H |
| Lump correction, MDM | A – C | A, E |

The results from these series of tests were used to perform the final activity: demonstration on real waste drums.

The capability of the CTEN system is measured against its performance compared to the WIPP's characterization requirements (DOE 1995). WIPP requires an assay uncertainty (due to systematic error, or bias) of 25% or less for SNM loadings at higher masses and at lower masses near the 100 nCi/g TRU/low-level waste (LLW) cutoff, and 50% or less for lower masses that are not close to the TRU/LLW cutoff. These limits are given quantitatively in Table 2 below, with mass ranges given for WG Pu. The 25% low mass limit is most significant for active mode assays, while the 25% upper mass limit is most significant for passive mode assays.

Table 2. WIPP accuracy requirements

| Waste activity (alpha Ci range) | Equivalent WG Pu mass range (g) | Accuracy requirement (+/- percentage) |
|---------------------------------|---------------------------------|---------------------------------------|
| >0.002 – 0.02 | >0.025 – .25 | 25% |
| >0.02 – 0.2 | >0.25 – 2.5 | 50% |
| >0.2 – 2 | >2.5 – 25 | 25% |
| >2 | >25 | 25% |

Treatment Performance

Passive Assay Accuracy

All current passive assay methods use either a drum-averaged or a multiplicity type of correction based on the triple coincidence rate, or both. The Los Alamos HENC uses a drum-averaged ("add-a-source") correction at lower mass loadings and a multiplicity technique at higher mass loadings. Existing PAN systems use a drum-averaged correction. There are no direct data on how other methods would perform on the surrogate drum set examined using CTEN, but it can be assumed that the multiplicity correction gives approximately the same results on all systems. Also, a theoretical limit can be set on performance of the drum-averaged technique. This is obtained by applying the average correction factor for a drum to every assay made in that drum. This is an optimistic limit for existing systems, which tend to do much worse than the limit with difficult matrices.

Table 3 shows the fraction of 180 passive assays of moderating drums that failed the WIPP assay accuracy requirements at the 25 and 50% error levels for uncorrected assays. Also shown, are the percentage of drums that failed for assays corrected for matrix effects using the multiplicity method; a drum-averaged correction based on CTEN's flux monitor count rates; the theoretical best possible drum-averaged correction; and the CTEN position-sensitive correction method. These are labeled "Uncorrected," "Multiplicity Correction," "CTEN Average Correction," "Best Possible Average Correction," and "CTEN Spatial Correction," respectively.

Table 3. Percentages of CTEN passive assays failing WIPP assay accuracy requirements

| WIPP maximum uncertainty | Uncorrected | Multiplicity correction | CTEN average correction | Best possible average correction | CTEN spatial correction |
|--------------------------|-------------|-------------------------|-------------------------|----------------------------------|-------------------------|
| 25% | 92.8% | 78.9% | 44.4% | 44.4% | 18.3% |
| 50% | 70.6% | 38.9% | 20.6% | 18.9% | 3.3% |



The CTEN average correction works nearly as well as the best accuracy that can be attained with a drum-averaged correction. And, as can be seen in the table, a factor of 2.4 and 5.7 times fewer assay failures are seen at the 25 and 50% levels, respectively, with the CTEN spatial correction, than with the best possible drum-averaged correction. In practical terms, this means that more difficult waste forms can be assayed using the CTEN passive mode than can be assayed using other passive counting methods, and that higher effective drum loadings can be certified for any given waste form. The latter is true because twice the uncertainty is added to the assayed mass value in determining whether a drum is over the 200 g fissile limit, so a smaller uncertainty allows higher plutonium loadings to stay below the limit.

Active Assay Accuracy

A common failing of the active (DDT) assay methods currently in use is that while their main application is in assaying small amounts of SNM (near the 100 nCi/g cutoff, or anywhere below the sensitivity levels of other methods), their matrix corrections only work well at high SNM loadings. This is not often clear in the literature, as performance characteristics are generally cited for larger SNM sources. In the older "second generation" DDT PAN systems, the active matrix correction is a drum-averaged correction derived from passive counting data, so good correction can be obtained only when assaying plutonium in amounts above approximately 0.5 g. Modern PAN systems, such as imaging passive active neutron (IPAN) and Active Passive Neutron Examination and Assay (APNEA), use tomographic image reconstructions to obtain spatial corrections. However, tomographic imaging in neutron assay systems requires exceptionally good counting statistics and can only work well at high SNM loadings (Estep November 1989). Moreover, the imaging techniques used in both IPAN and APNEA are for specific matrix types only; no method has been devised as yet to "interpolate" the images to intermediate matrix types or to general types, as is done automatically in CTEN. At lower SNM loadings, these systems must fall back on drum-averaged techniques.

As with the CTEN passive spatial correction, the CTEN active assay matrix correction uses two robust active indicators of average positional effects to obtain spatial corrections that give significantly better corrections than a drum-averaged approach. These are the ratio of top-to-bottom detector counts (which gives an average height indication) and the sum of the squared deviations of the individual detector packages over the 12 measurement angles (which gives an average depth indication). Both quantities have low error amplification factors and are measured with high sensitivity in the active assay, and so give good spatial corrections down to approximately 20 mg of plutonium (this is below the lowest mass listed in Table 2).

The relative merits of the CTEN passive and active spatial corrections compared with tomographic imaging and with drum-averaged corrections will be discussed in an upcoming report (Estep, Melton, and Miko November 1998). Preliminary data are presented in Table 4, which shows the fraction of 460 active assays of moderating and absorbing drums that failed the WIPP accuracy requirements for the uncorrected case, the best drum-averaged correction, and the CTEN active spatial correction. As can be seen in the table, the spatial correction results in a factor of 6.8 fewer assays falling outside the important 25% WIPP uncertainty limits than with the best possible drum averaged correction. No cases fell outside the 50% uncertainty limit versus 0.5% for the best possible drum-averaged correction. Note that the 25% uncertainty limit applies when sorting TRU waste from low-level waste.

Table 4. Percentages of CTEN active assays failing WIPP assay accuracy requirements

| WIPP maximum uncertainty | Uncorrected | Best possible average correction | CTEN spatial correction |
|--------------------------|-------------|----------------------------------|-------------------------|
| 25% | 84.6% | 23.9% | 3.5% |
| 50% | 59.5% | 0.5% | 0.0% |

Lump Correction Performance

The lump correction works poorly on WG Pu because of an inopportune resonance in the neutron absorption cross-section in ²³⁹Pu, and so is effectively limited to use with highly enriched uranium (HEU). This is not a drawback; passive neutron counting gives good assays of lumpy WG Pu, but cannot assay HEU, so the CTEN lump correction fills a gap in current NDA capability. The ⁴He detectors used in the epithermal interrogation have a poor sensitivity, so at least 3 g of HEU (finely divided equivalent) are



needed to perform reliable corrections. Also, the presence of hydrogen in the drum matrix interferes with the correction, which complicates the analysis and causes degraded sensitivity in moderating drums. Above a hydrogen content of approximately 0.01 g/cm³, the HEU lump correction will not work reliably.

It is recommended that the lump correction not be routinely incorporated into assay results. The deployed CTEN software will offer lump identification and correction as an option for those situations where it would be useful. When enabled, the software will report the likelihood that the SNM is in lumpy form and will suggest a correction factor without applying the correction to the assayed mass value.

Real Waste Demonstration

Table 5 lists the assay results for the real waste demonstration using four different active analysis methods (highlighted columns): no correction, corrections with no positional correlations, corrections with positional correlations based on CTEN indicators, and corrections with forced positional correction based on TGS images (Estep et al. June 1998). The first column uses an empty drum calibration factor and represents the measurement with no corrections. The remaining columns all have matrix corrections derived from CTEN data with varying knowledge of the source position included in the correction. The TGS scans were useful in determining how well CTEN's gross position indicators worked in locating material. The goal was not to pinpoint the contamination, but to determine whether material is located in the inner or outer radial regions, and top, middle, or bottom sections. By viewing TGS scans it was determined that this limited imaging goal was indeed accomplished. Using this coarse neutron imaging knowledge, the range of possible calibration factors was reduced, resulting in a significant improvement in measurement accuracy. In the far right column of Table 5 are the CTEN passive assay results, using matrix corrections based on active multiplicity analysis (Hollas et al. 1997). CTEN passive assay values with asterisks had high singles neutron rates. The CTEN assay results are compared with TGS assay results and with the previously reported PNCC and Segmented Gamma Scanner (SGS) results. In the remainder of this section, the values obtained are reported along with several noteworthy items from the demonstration.

- Drum #56130 Contains 243Cm – Although labeled as containing only Pu and previously assayed by PNCC as containing 31.8 g of Pu, anomalous values of the active and passive CTEN assay results, as well as various other indicators, suggested that drum #56130 might contain Cm rather than Pu. This was verified by examination of the cumulative gamma-ray spectrum of that drum from the TGS assay, in which the two strongest peaks were found to be from 243Cm. The TGS and SGS were unable to observe Pu peaks, although assay of Pu at less than about 500 mg is difficult. In spite of the presence of the Cm, the CTEN was able to make a determination of the Pu content using the active mode.
- Agreement between CTEN and TGS – Several trends were identified in Table 5. A significant trend is that when a comparison can be made, the CTEN results using positional corrections agree with the TGS assay results more than when the positional corrections are excluded. This can be understood as a heterogeneity effect in which the SNM is concentrated in one place where the matrix interference is more or less severe than the drum-averaged value. The simple drum-averaged analysis in such cases will either undercorrect or overcorrect. Only slightly better agreement was obtained when the positional parameters in the active analysis were "manually" forced to agree with the Pu images obtained from the TGS assay. This implies that the simple spatial correction technique used in CTEN is approximately correct.
- The case of drum #55925 illustrates that for significant errors to occur in CTEN active assays, fairly unusual circumstances must prevail. In particular, the matrix must be nonbenign and heterogeneous, with a large fraction of the SNM positioned in such a way that the matrix interference is either much less or much more than is probable. Specifically, this drum was approximately three-fourths full with a fairly homogeneous matrix of NaCl. There was a concentration of Pu near the surface of the matrix, where it was only half shielded by the matrix. This geometry results in overcorrection of the mass value, as the analysis assumes that the Pu is embedded in the matrix and not sitting on top of the matrix in the open. It is expected that such drums will be uncommon in the overall waste stream.

Key System Parameters

Isotopic data on the waste must be known before completing the CTEN measurement.



Limitations/Potential Problems

- The current CTEN hardware configuration limits the container size to 55-gallon drums or 83-gallon overpacks. A boxed waste assay system, based on the CTEN technology, is currently being fabricated at LANL. The box assay system will undergo calibration and testing in fiscal year (FY)-99.
- Isotopic information of the waste must be known either through process knowledge or gamma spectrometry.
- Passive counting does not work well with high singles or uncorrelated neutron count rates [for example Americium produces high (α, n) levels in containers]. This would be a problem for sludge type wastes generated from reprocessing, e.g., at Rocky Flats Environmental Technology Site (RFETS).

Table 5. Real Waste Assay Results

| Waste ID | Matrix Material | Drum Weight (kg) | Declared Value Total Pu (mg) | CTEN Active Measurement Results | | | | TGS Results Total Pu (mg) | CTEN Passive Matrix Corrected from Active Total Pu (mg) |
|---|----------------------|------------------|------------------------------|--|--------------------------------------|--|---------------------------------------|---------------------------|---|
| | | | | No Matrix or Position Correction Total Pu (mg) | Matrix Corrected | | | | |
| | | | | | No Position Correction Total Pu (mg) | CTEN Position Correction Total Pu (mg) | TGS Position Correction Total Pu (mg) | | |
| 56154 | Plastic/Kimwipes | 11.77 | 0 | 8.28 | 10.65 | 10.66** | - | - | |
| 56162 | Rubber | 26.53 | 0 | 10.79 | 13.88 | 14.04** | - | - | |
| 56156 | Plastic/Kimwipes | 14.9 | 1 | 75.56 | 1.50 | 14.00** | - | - | |
| 56061 | Plastic/Kimwipes | 18.2 | 8 | 34.01 | 39.10 | 38.50 | - | - | |
| 56158 | Plastic/Kimwipes | 22.3 | 158 | 56.36 | 81.90 | 94.50 | - | - | |
| 56078 | Plastic/Kimwipes | 16.55 | 168 | 57.61 | 74.10 | 77.80 | - | - | |
| 56159 | Plastic/Kimwipes | 15.6 | 194 | 10.29 | 12.90 | 12.4** | - | - | |
| 56144 | Graphite | 15.33 | 218 | 136.68 | 126.90 | 126.30 | - | - | |
| Waste drums listed above have declared masses that are below the nominal detection limits for SGS, TGS, and NCC | | | | Total Pu (g) | Total Pu (g) | Total Pu (g) | Total Pu (g) | Total Pu (g) | Total Pu (g) |
| 56132 | Non-Actinide metal | 193.02 | 1.529 | 0.331 | 1.0950 | 1.4870 | *** | *** | 2.46 |
| 56031 | HEPA | 31.61 | 3.898 | 1.265 | 3.1130 | 3.4070 | 3.4070 | 3.7060 | 4.19 |
| 56086 | Paper, Wood, Plastic | 21.89 | 4.36 | 2.519 | 5.7200 | 5.3230 | 5.2840 | 4.9760 | 4.02 |
| 56147 | Non-Actinide metal | 44.5 | 10.902 | 0.6573 | 0.8452 | 0.8887 | 0.8752 | 0.6548 | 10.97 |
| 56151 | Paper, wood, rubber | 12.31 | 11.726 | 18.431 | 17.1050 | 16.578 | 15.840 | 10.960 | *17.75 |
| 56140 | Glass | 35.2 | 13.641 | 3.936 | 8.553 | 8.490 | 7.841 | 8.171 | 13.11 |
| 56089 | Non-Actinide metal | 40.8 | 19.607 | 4.062 | 15.222 | 15.503 | 12.059 | 10.088 | *19.92 |
| 56087 | Plastic/Kimwipes | 28.75 | 20.774 | 14.124 | 21.020 | 20.673 | 20.286 | 21.792 | *24.25 |
| 56088 | Non-Actinide metal | 45.3 | 21.473 | 15.72 | 22.842 | 25.610 | 25.387 | 16.667 | 22.04 |
| 56130 | Glass | 84.94 | 31.832 | 2.787 | 2.033 | 1.723 | 1.812 | 1.612 | *48.84 |
| 56138 | Plastic/Kimwipes | 21.48 | 31.967 | 37.577 | 27.416 | 31.197 | 28.248 | 25.862 | 40.07 |
| 56072 | Non-Actinide metal | 54.25 | 41.496 | 16.408 | 32.293 | 39.708 | 24.631 | 27.868 | 40.7 |
| 56118 | Plastic/Kimwipes | 22.24 | 44.792 | 33.951 | 38.978 | 37.937 | 39.409 | 45.406 | 39.17 |
| 56123 | Non-Actinide metal | 100.43 | 96.01 | 25.545 | 62.852 | 79.885 | 70.270 | 68.802 | 86.17 |
| 56098 | Plastic/Kimwipes | 48.65 | 120.12 | 59.208 | 121.907 | 101.530 | 92.263 | 114.511 | 138.24 |
| 56038 | Salt/Chloride/Oxide | 70.31 | 172.81 | 59.704 | 88.857 | 87.320 | 91.316 | 100.073 | *187.12 |
| 55925 | Salt/Chloride | 118.08 | 178.51 | 101.859 | 209.720 | 187.127 | 173.178 | 107.101 | *246.43 |

* These passive assays had high singles neutron rates.

** Low-level waste (<100 nCi/g).

*** Drum weight out of TGS prototype's range.



SECTION 4

TECHNOLOGY APPLICABILITY AND ALTERNATIVES

Baseline and Competing Technologies

NDA of waste containers using neutron methods are available both commercially and at the DOE laboratories. Two common instruments that are commercially available are the DDT/PAN and HENC systems.

The DDT/PAN method is discussed briefly throughout this report since the CTEN is basically an improved DDT/PAN system. To summarize the DDT/PAN technology, it is used to measure the fissile content (usually ^{239}Pu , ^{241}Pu , or ^{235}U) in radioactive waste containers. This method uses thermalized neutrons from a 14-MeV pulsed source to irradiate a drum. Prompt neutrons from induced fissions are detected and thus provide a direct measure of the fissile content of the drum. In addition to interrogation of the container with thermal neutrons (active mode), DDT/PAN instruments also perform passive coincidence measurements (passive mode). Spontaneously fissioning isotopes such as ^{252}Cf and the even isotopes of plutonium can be measured using this technique.

The HENC system is a passive neutron coincidence counter. This technique measures spontaneous fission neutrons from the three even isotopes of Pu. The HENC has a detection efficiency of 30% and increased shielding to reduce background interference. The system's detection level is on the order of 1.6 mg of ^{240}Pu -effective in a 1,000 second count time. The HENC also incorporates enhanced correction and analysis techniques such as Add-A-Source and multiplicity counting are added to provide assay accuracy.

PAN systems at INEEL, Pacific Northwest National Laboratory, LANL, and RFETS have participated in the PDP. British Nuclear Fuels, Limited (BNFL) instruments PAN have also been demonstrated in Cycles 3 and 4 of the PDP. The HENC system recently participated in the Capabilities Evaluation Project (CEP) at the INEEL. This project was funded by the MWFA and the Characterization, Monitoring, and Sensor Technology Crosscut Area to evaluate the capability of existing mobile assay systems. Performance data from the PDP and CEP projects can be found in References (PDP Scoring Report November 1996; PDP Scoring Report May 1997; Becker September 1998; and Becker December 1998).

Technology Applicability

Active neutron assay is the only technique that can assay TRU waste near the TRU/low-level cutoff in a majority of waste forms, but previous active neutron systems do not meet WIPP accuracy requirements with many common waste forms. The CTEN's improved active assay accuracy at low SNM mass loadings allows certification of a significant number of drums that cannot be certified by any other technique.

Technology Status and Maturity

The CTEN is a technically mature system; no further research or development is required for deployment. However, deployment of the CTEN does require that the software be rewritten and documented to meet quality assurance standards, the user interface be simplified, and the system participate in formalized testing and validation (e.g., PDP). These activities will be completed during the deployment of the CTEN to LANL in FY-99.

Patents/Commercialization/Sponsor

LANL is working to commercialize the technology. The technology has not been patented and has been made available for use by any DOE and non-DOE interested parties.



SECTION 5

COST

Methodology

The following costs are based on siting costs at LANL and cost estimates of commercially-available DDT/PAN systems (Estep, ASTD Proposal).

Cost Analysis and Conclusions

Two deployment scenarios are outlined in this section: deployment of a newly fabricated system and upgrade of an existing DDT/PAN system.

New System Costs

The price of a commercially-available DDT/PAN system is approximately \$1,000,000. The Principal Investigator (PI) estimated that the delivered cost of a CTEN from a vendor such as Canberra Industries or BNFL/Pajarito would be approximately 1,000,000 to \$1,200,000. The PI estimated that the cost for LANL to fabricate another CTEN system would be roughly equivalent.

Additional costs to install a new system at a DOE site include facility siting and certification. Siting costs at LANL's CST-7 area are relatively low as compared to some other DOE facilities. There are several reasons for this: CST-7 is a low-security area and they have experienced NDA people who are actively working with similar DDT systems. However, it should be noted that some of the work completed for the LANL deployment may be transferable to other sites; for example, characterization of total uncertainty. The cost estimates for deployment at LANL include the following:

- PDP certification: ~ \$70,000,
- installation and certification: \$790,000,
- operating costs (manpower): \$500,000/year,
- facility costs (operating or idle): \$133,000/year.

Upgrade of an Existing System Costs

An existing system can be upgraded to add list-mode counting with the latest PCI-bus PATRM. This will improve multiplicity analysis of the data. However, there are several attributes of the CTEN that this upgrade will not include: epithermal interrogation and limited improvement to spatial resolution. The base price for the hardware and software would be approximately \$50,000. The time for installation and support costs would be approximately \$200,000. PDP certification and operating and facility costs would be the same as for the new system costs.



SECTION 6

REGULATORY AND POLICY ISSUES

This section presents current and anticipated regulatory requirements of an NDA technology end user to meet site and disposal facilities characterization requirements. The specific regulatory requirements and their associated issues that pertained to the CTEN development effort are also described.

This section also presents an analysis performed by the MWFA that assesses the various risks involved with deployment of the CTEN and pertinent stakeholder responses to the technology's application.

Regulatory Considerations

The objective of using a CTEN system is to characterize all radioisotopes within a mixed waste container nondestructively (without opening the container). Major regulatory requirements, including permit/license requirements, for implementation of this technology are expected to include:

- National Environmental Policy Act (NEPA) review for implementation at federal facilities. At DOE facilities, this includes an initial environmental checklist that is used to assist in determining if a more detailed environmental assessment or environmental impact statement is required.
- A radioactive material license from the Nuclear Regulatory Commission (NRC) or its applicable agreement state for non-DOE facilities or for DOE facilities expected to be regulated by the NRC or the agreement state.
- If the CTEN system is to be used to ensure regulatory compliance, review and possible approval to the methodology would be needed from the applicable regulatory authority.

Safety, Risks, Benefits, and Community Reaction

Eight risk areas were evaluated and assessed independently. These risk values for MWFA developed technologies have been derived from the eight top-level requirements defined in the MWFA Systems Requirements Document [Idaho National Engineering Laboratory (INEL) 1997]. The eight areas evaluated for level of risk are: (1) ease of permitting, (2) technical correctness, (3) level of safe operability, (4) technical completeness (i.e., ready to use), (5) timely to meet treatment schedules, (6) acceptability to stakeholders, (7) cost-effectiveness to use, and (8) committed sponsorship. A complete description of the methodology and a detailed definition of each risk element, the event scenario, and the basis for assigning consequences and probability factors are included in Appendix C.

Permittable: The risk category is rated as low and improbable that a permit application will be rejected. This is not a treatment process, therefore a permit to operate is not required. A site must certify an instrument to use in characterizing TRU waste.

Complete: The risk category is rated as medium and improbable that additional engineering is required to allow the instrument to be incorporated into a system. The system has not yet formally participated in the National TRU Program's PDP. There is a limited amount of testing that would be required of a system that will be characterizing waste for disposal at the WIPP.

Acceptable: The risk category for acceptable is rated as low and improbable that a Native American Tribe or public interest group would resist that implementation of the CTEN technology at a DOE site.

Timely: The risk category is rated as low and improbable that the technology will not be available by Site Treatment Plan or Consent Order dates. The CTEN system development has been completed and information is available for construction of a new system.

Cost: The risk category is rated as medium and improbable that the operational costs will be higher than expected. The cost analysis is based on the characterization of a large volume of waste. Even a minor difference in cost could affect a site if large quantities of waste were targeted.



Sponsored: This category is listed as low and unlikely that no end-user or commercial entity selects the technology for implementation. The CTEN system will be deployed at LANL in FY-99.

Correct: This category is listed as medium and improbable that the technology will not be applicable to the target waste. Multiple waste types were measured using this instrument to identify the applicable target streams.

Safe: This category is listed as low and improbable that system failure will adversely impact the health and/or safety of a collocated worker, the environment, or a member of the public. No hazardous materials will be added or generated during the measurement. Radiological hazards should be no different than commonly accepted medical techniques, i.e., cat scans.

The MWFA Tribal and Public Involvement Resource Team reviewed stakeholder issues and concerns related to characterization of mixed wastes. The risk to the community is very low. In general, the public has limited familiarity with nondestructive assay systems such as the CTEN, but would be expected to support it as an improvement. The issues of concern to the public are discussed:

- Community Safety
- There is no adverse safety impact to the community
- Potential Socioeconomic Impacts and Community Perceptions
- No socioeconomic impacts are anticipated
- No adverse public or tribal input regarding the CTEN technology was received
- Comparisons of this technology to x-ray systems currently in use (such as those in dentists offices or hospitals) may also reduce the anxieties associated with the sources
- Benefits
- The improved accuracy and reliability of the system can provide the public with increased confidence in assay data
- There will be no potential to release contaminants to the environment.



SECTION 7

LESSONS LEARNED

Implementation Considerations

Owners of mixed waste and potential technology end users have several choices for waste characterization. These choices include the CTEN technology, a DDT/PAN system or any other neutron or gamma-based NDA system. It is recognized that an end user may need to use a suite of technologies to address their waste assay needs. Factors that should be considered when evaluating the use of this or any NDA technology include:

- System assay times – Technology operations should not affect site schedules.
- Preparation time to start assay – Technology operations should not affect site schedules.
- Ease of operation/maintenance – Simplicity of operation and ease of maintainability, e.g., time required for infrastructure system setup and takedown (stairways, shielding, utility/data connections, etc.), number of operators, systems simple to operate, etc.

Design Issues

CTEN has no unresolved design issues.

Technology Limitations and Needs for Future Development

There are two limitations to any DDT/PAN system, including the CTEN. These limitations include:

- Waste Container Isotopics. Isotopic knowledge of the waste is required to generate the characterization data required by WIPP. The isotopic information can be obtained in two ways: (1) by gamma spectrometry for containers with >1 g Pu and (2) by process knowledge or destructive analysis for containers with <1 g Pu.

NDA of Containers with Sources of (α,n) . Passive counting does not work well with high singles or uncorrelated neutron count rates.



APPENDIX A

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APPENDIX B

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APPENDIX C

RISK ASSESSMENT METHODOLOGY

Risk has been measured for eight of the system requirements as defined in the MWFA Systems Requirements Document.

Technically Correct (Correct)

The MWFA shall deliver treatment technologies that are technically correct. Operable treatment systems shall be able to: (1) treat target waste streams identified in Federal Facility Compliance Act (FFCA) Site Treatment Plans (STPs) and (2) treat wastes to meet Environmental Protection Agency (EPA) treatment standards (and Toxic Substance Control Act or state-regulated treatment standards, where applicable) and comply with the disposal facility Waste Acceptance Criteria.

Technically Complete (**Complete**)

Treatment technologies delivered by the MWFA shall be demonstrated to function as described, and shall be described in sufficient detail so that they may be incorporated into a detailed system design of a mixed low-level or mixed transuranic waste treatment system without further development.

Acceptable to Stakeholders (**Acceptable**)

The MWFA shall deliver mixed waste treatment technologies that are acceptable to the stakeholders.

Note: The term "stakeholders" means all those who have an interest in the outcome of the MWFA program except the DOE and DOE contractors who have a direct and immediate interest or involvement in the MWFA. Stakeholders include: tribal governments, members of the public, federal, state, and local agencies, universities, and industry.

Acceptable to an End User (**Sponsored**)

The MWFA shall deliver mixed waste treatment technologies to users committed to pursuing the use of those treatment technologies in mixed waste treatment systems.

Permittable

The MWFA shall deliver mixed waste treatment technologies along with sufficient data to show that there are no probable technical reasons to prevent receiving a permit to implement the technology in an operational treatment system. The permit process will be facilitated by involvement with national regulatory organizations such as NTW on Mixed Waste Treatment and Interstate Technology and Regulatory Cooperation Subgroup. This will include working with the regulators to improve technologies and/or a facility's ability to obtain a permit.

Safe

The MWFA shall deliver mixed waste treatment technologies that can be incorporated into a treatment system and safely operated.

Timely

The MWFA shall deliver mixed waste treatment technologies to enable treatment systems to be designed, built, and operated in time to meet treatment schedules in the FFCA STPs and negotiated in Consent Orders.



Cost

The “delta” refers to the cost of implementation by an end user when compared to the cost analysis included in the ITSR. The more closely the cost of implementation compares with cost as reported in the ITSRs, the smaller the consequence to the end user of the technology.

Each of the eight system requirements will be addressed independently. Events that can lead to negative consequences relative to implementation of a technology will be identified and assigned to each system requirement. These events will be referred to as “risk factors.” Each technology will be evaluated independently and relative values for consequences and probability will be assigned to each of the events. Criteria have been defined for each risk category to allow the user to, as quantitatively as possible, determine the probability and consequence measures to be applied for determination of risk.

Permittable

Permit application is rejected based on regulations that became effective after development of the technology.

The consequences of this scenario will be:

- Low if Treatment process is simple.
- Medium if Treatment process is complex.
- High if Treatment process is highly complex.

The probability of this scenario occurring will be:

- Improbable if An applicable permit has been received.
- Unlikely if Regulators have maintained interaction with developers on this technology during development and demonstration.
- Likely if A permit application has already been rejected for this technology.

Complete

Technology is insufficiently mature to incorporate into a system without additional engineering data.

The consequences of this scenario will be:

- Low if Technology can be deployed without the need for additional testing.
- Medium if Technology can be deployed with limited additional testing and documentation.
- High if Technology requires significant additional development and/or testing to deploy.

The probability of this scenario occurring will be:

- Improbable if Technology successfully meets Stage 5 requirements for full system functionality and has successfully conducted a treatability study.
- Unlikely if Technology successfully meets Stage 5 requirements for full system functionality and has conducted successful demonstration(s) with surrogate wastes.
- Likely if Technology successfully meets Stage 5 requirements for full system functionality, but demonstration/testing program is incomplete.



Acceptable

Native American Tribes and/or public interest groups resist implementation of the technology at DOE sites.

The consequences of this scenario will be:

- | | |
|-----------|--|
| Low if | Concerns can be addressed by providing additional information about the technology's performance. |
| Medium if | Concerns center on the performance of the technology; relatively minor modifications to the technology can address the needs and concerns. |
| High if | Major modifications to the technology are required to address concerns about the performance and ability to solve the problem. |

The probability of this scenario will be:

- | | |
|---------------|--|
| Improbable if | The affected Tribes and public perceive implementation of the technology as resolving an important problem at their site with minimal or no impact to their quality of life, or have not expressed any concerns. |
| Unlikely if | The affected Tribes and public perceive implementation of the technology as solving an important problem but having a negative impact on the quality of life. |
| Likely if | The affected Tribes and public perceive implementation of the technology will not solve an important problem at the site and is perceived to have significant negative impact on the quality of life. |

Timely

The technology is not available for implementation by the STP or Consent Order date.

The consequences of this scenario will be:

- | | |
|-----------|--|
| Low if | Delay in the availability of the technology will not result in missing a milestone in a Consent Order. |
| Medium if | Need dates for the Consent Order can be renegotiated to accommodate the delay in availability of the technology. |
| High if | Unavailability of the technology results in missing key milestones in Consent Orders at multiple sites. |

The probability of this scenario will be:

- | | |
|---------------|---|
| Improbable if | Technology development/implementation activities are completed within end user schedules. |
| Unlikely if | Need dates identified accommodate any minor delays in technology development activities. |
| Likely if | Technology does not meet end-user schedules. |



Cost

Operational costs are higher than projected.

The consequences of this scenario will be:

- Low if Volume of the targeted waste is low.
- Medium if Volume of the targeted waste is fairly small.
- High if Volume of the targeted waste is very large.

The probability of this scenario will be:

- Improbable if Projections of the technology's cost is based on data from multiple campaigns.
- Unlikely if Projections of the technology's cost is based on data from only one campaign.
- Likely if No actual cost data for the technology on the targeted waste exists.

Sponsored

No end-user or commercial entity selects the technology for implementation.

The consequences of this scenario will be:

- Low if Multiple data sets detailing the technology's performance on targeted waste are available.
- Medium if Only limited data are available detailing the technology's performance on targeted waste.
- High if Data are not available detailing the technology's performance on the targeted waste.

The probability of this scenario will be:

- Improbable if Multiple licensing agreements or financial commitments have been made.
- Unlikely if A single licensing agreement or financial commitment for the technology has been made.
- Likely if No commitments have been made or interest shown in the use of the technology.

Correct

Operable treatment systems, which incorporate this technology, are not applicable to target wastes.

The consequences of this scenario will be:

- Low if Volume of targeted waste to be treated is low.
- Medium if Volume of targeted waste to be treated is fairly small.
- High if Volume of targeted waste to be treated is very large.

The probability of this scenario will be:

- Improbable if Technology developed was tested against multiple waste types.
- Unlikely if Technology developed was tested against only one waste type.
- Likely if Technology developed was not tested against targeted waste type.



Safe

System failure adversely impacts the health and/or safety of a collocated worker, the environment, or a member of the public.

The consequences of this scenario will be:

- | | |
|-----------|--|
| Low if | Hazardous constituents added or generated by the system are less than the reportable quantities shown in 40 CFR 302.4 and 40 CFR 355, Appendix A. |
| Medium if | Nominal reportable quantities of hazardous constituents shown in 40 CFR 302.4 and 40 CFR 355, Appendix A, are added or generated by the system. |
| High if | Hazardous constituents in quantities 10 times or greater than those listed in 40 CFR 302.4 and 40 CFR 355, Appendix A, are added or generated by the system. |

The probability of this scenario will be:

- | | |
|---------------|---|
| Improbable if | System is a benign process, difficult to combust with no natural gas or fuel sources present. |
| Unlikely if | System is a moderately energetic process with natural gas or fuel sources present. |
| Likely if | System is an energetic system (high temperature and/or pressure); large amounts of flammables or pyrophorics. |



APPENDIX D

ACRONYMS

| | |
|--------|---|
| APNEA | Active Passive Neutron Examination and Assay |
| BNFL | British Nuclear Fuels, Limited |
| CEP | Capabilities Evaluation Project |
| Cf | Californium |
| Cm | curium |
| CTEN | Combined Thermal Epithermal Neutron |
| DDT | differential dieaway technique |
| DOE | Department of Energy |
| EPA | Environmental Protection Agency |
| FFCA | Federal Facility Compliance Act |
| FY | Fiscal Year |
| He | helium |
| HENC | High Efficiency Neutron Counter |
| HEPA | high-efficiency particulate air |
| HEU | highly enriched uranium |
| INEEL | Idaho National Engineering and Environmental Laboratory |
| INEL | Idaho National Engineering Laboratory |
| IPAN | imaging passive active neutron |
| ITSR | Innovative Technology Summary Report |
| LANL | Los Alamos National Laboratory |
| LLW | low-level waste |
| LMITCO | Lockheed Martin Idaho Technologies Company |
| MDM | minimum detectable mass |
| MLLW | mixed low-level waste |
| MTRU | mixed transuranic waste |
| MWFA | Mixed Waste Focus Area |
| NDA | nondestructive assay |
| NDE | nondestructive examination |
| NEPA | National Environmental Policy Act |
| NRC | Nuclear Regulatory Commission |
| NTW | National Technical Workgroup |
| OST | Office of Science and Technology |
| PAN | passive active neutron |
| PATRM | Pulse-Arrival Time Recording Module |
| PDP | Performance Demonstration Program |
| PI | Principal Investigator |
| PNCC | passive neutron coincidence counter |
| Pu | plutonium |
| RFETS | Rocky Flats Environmental Technology Site |
| SGS | Segmented Gamma Scanner |
| SNM | special nuclear material |
| STP | Site Treatment Plan |
| TGS | Tomographic Gamma Scanner |
| TRU | transuranic |
| U | uranium |
| WG | weapons grade |
| WIPP | Waste Isolation Pilot Plant |

