A REVIEW OF REAL-TIME RADIOGRAPHY OF REMOTE-HANDLED TRU WASTE

July 2006



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ISO-2 Project Carlsbad, NM

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ACRONYMS

AK ASTM	acceptable knowledge American Society for Testing Materials
CBFO CH Ci Cs-137 Co	Carlsbad Field Office contact-handled curie(s) cesium 137 cobalt
DOE	Department of Energy
Gd	gadolinium
HWFP	Hazardous Waste Facility Permit
INL Ir	Idaho National Laboratory iridium
KeV	thousand electron volt(s)
LANL	Los Alamos (National Laboratory)
MeV mrem	million electron volt(s) milliRoengten equivalent man
NDA NDE NMED	non-destructive analysis non-destructive examination New Mexico Environment Department
0	oxygen
Pb PC	lead (plumbum) personal computer
rem RH RTR	Roentgen equivalent man remote-handled real-time radiography
S SIT SRS	sulfur Silicon Intensifier Target Savannah River Site
TRU	transuranic
UR	unclassified report
VE	visual examination
WAC WIPP	waste acceptance criteria Waste Isolation Pilot Plant

A REVIEW OF REAL-TIME RADIOGRAPHY OF RH TRU WASTE — July 2006

I. SCOPE AND PURPOSE

The purpose of this report is to review available information relating to the effectiveness of real-time radiography (RTR) for characterization of remote-handled (RH) transuranic (TRU) waste destined for disposal at the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico. This report is based on the following: a review of the documentation and discussion of RTR presented in the proposed modification to the Hazardous Waste Treatment Facility permit and the Compliance Recertification Application for the WIPP; previous studies conducted for DOE concerning the use of RTR for RH TRU waste; and an independent peer review of the technology. Individuals designated by DOE as knowledgeable in RTR have also provided supplemental information. This report does not compare the relative merits of RTR with other methods of characterization such as acceptable knowledge (AK) and visual examination (VE). Though the latter two methods are expected to be employed on over 90% of the RH TRU waste volume sent to the WIPP, it is anticipated that RTR will be used to characterize the small fraction of containers for which AK is insufficient and/or in cases when high dose rates prohibit the use of VE.

In the field of nuclear chemistry, x-rays and gamma rays are defined by the method of production.^{1,2} X-rays are produced by the electrons of an atom returning to ground state from an excited state, or by electrons that are deflected from their original path. Gamma rays result from a nucleus of an atom returning to ground state from an excited state. By contrast, physicists distinguish between x-rays and gamma rays only by their energies; not by the method of their production.³ In this report we will use the definitions that are common to the field of nuclear chemistry—those used in the cited references and that have been historically used in radiochemistry in the United States of America. (We will not use SI [Systeme International d'Unites] units.)

II. BACKGROUND

The TRU waste currently being emplaced at the WIPP is contact-handled (CH). Future plans include emplacing RH TRU waste in the repository. The classification of TRU waste as CH or RH depends on the dose rate measured at the surface of the waste container. TRU waste with a dose rate exceeding 200 millirems (mrems) per hour and less than 1000 rems per hour at the container surface is defined as RH TRU waste. No more than 5% of the RH TRU waste by volume to be disposed in the WIPP is allowed to exceed a surface dose rate of 100 rems per hour. No waste drum with a surface dose rate in excess of 1000 rems per hour can be accepted for disposal at the WIPP. All waste, whether CH or RH, must be characterized and must meet the Waste Acceptance Criteria (WAC) prior to emplacement in the repository. The current Hazardous Waste Facility Permit (HWFP) requires that all WIPP waste containers undergo VE, AK, or RTR in order to confirm the absence of prohibited items such as liquids and pressurized containers. RTR will only be used when AK is not sufficient and when higher dose rates of RH TRU waste prevent the use of VE.

RTR is a non-destructive examination technique that uses x-rays or electromagnetic radiation produced by Bremsstrahlung to create images of the interior of waste containers. The "real-time" component is crucial, as it allows the radiographer to examine an image of the contents of a waste container and make an immediate determination as to whether or not prohibited items are present. While the RTR technique is not without limitations (discussed below), it does offer a view of the inside of TRU waste containers while providing protection for the operator in the infrequent cases when VE methods cannot be used. Materials with higher densities within the TRU waste containers will require more intense or energetic xrays to create images of those materials. In summary, the RTR equipment enables an operator to adjust both the energy and intensity of the x-rays, providing the operator independent degrees of freedom and the flexibility to identify a container's contents while simultaneously interrogating its interior.

III. SUMMARY OF FINDINGS

A report entitled *Real Time X-Radiography for Examination of Remotely Handled Radiographic Objects*,⁴ authored by B. W. Brown and C. R. Mikesell, describes several experiments conducted on two simulated waste drums that had the capability for the experimenters to insert radioactive sources within the drums. Three instruments interrogated these drums: a 420 KeV Phillips x-ray source, a six MeV portable linear accelerator (Minac-6), and a nine MeV stationary linear accelerator (Linatron 3000A). Results from the experiments pertiment to this report are presented below:

One drum (Drum #1) contained a variety of objects and materials of different densities that typically comprise CH waste, including plastic bottles containing water and ranging in size from 2 ounces to 1 pint; an aerosol spray-paint can; plastic bottles containing various materials such as metallic chips and floor sweepings; a steel object approximately 2 inches in diameter and 9 ½ inches long; folded cardboard, and a fiberpack container with a steel lid that contained several large pieces of a graphite-like material.

The second drum (Drum #2) was half-filled with a synthetic sludge. (The composition of the sludge was not given.) Within the sludge were several items: a 4- or 5-ounce plastic bottle containing water; 2 golf ball-size pieces of a Teflon-like material; an aluminum plate approximately 6 inches by 12 inches; a piece of threaded steel rod measuring 3/8 of an inch by 10 inches; and a piece of stainless steel angle iron. In addition to the above items, approximately 1 quart of water was placed in a puddle on top of the sludge.

Each drum was fitted with a 3-inch-diameter tube that was closed on the bottom end and located on the centerline of the drum. This tube passed through the lid of the drum, which was attached to the tube by a locking nut. The purpose of the tube was to contain the radioactive sources that were placed in the drums for some of the experiments.

The radioactive sources used in these experiments were 60 Co (28 Ci) and 192 Ir (100 Ci). The gamma radiation emitted by 60 Co has energies of 1.173 MeV and 1.332 MeV. The 192 Ir has four gamma rays ranging in energy from 0.296 MeV to 0.468 Mev.

III.A. 420 KeV X-Ray System Experiments

Drum #1 was interrogated using the 420 KeV x-ray system. The image quality was very satisfactory. When the drum was scanned a second time with the ⁶⁰Co source in the viewing area of the detector, the image of the ⁶⁰Co capsule was "sharp and detailed." No loss of image quality was reported when the ⁶⁰Co source was present. Drum #2 could not be satisfactorily interrogated due to the inability of the x-ray

source to fully penetrate the drum. The authors of the report considered the ⁶⁰Co source to represent an "intermediate-level radioactive waste." The dose rate at the surface of either drum was not reported, although it appears from subsequent studies discussed below to be in excess of 1000 R/hour.

In conclusion, the 420 KeV x-ray system adequately imaged the contents of Drum #1, which did not contain sludge; but this system was incapable of imaging the contents of Drum #2, which did contain sludge.

III.B. 6 MeV "Minac-6" Electron Linear Accelerator System Experiments

Initially, Drum #1 (without the radioactive sources) was interrogated, and with the exception of the folded cardboard, floor sweepings, and the metallic chips, all other items were satisfactorily imaged. Liquids in the drums were readily discernable. Plastic bags used to contain items were visible only when the layers of plastic overlapped.

In a second experiment, the two radioactive ⁶⁰Co and ¹⁹²Ir sources were placed in the field of view and the drum was then interrogated. The 2-ounce plastic bottle was visible, and the level of the liquid was discernable. The dose rate from the accelerator beam was between 20 and 25 R/min. The ratio of accelerator photons to the photons from the radioactive ⁶⁰Co and ¹⁹²Ir was between 5 and 15. Throughout the range of ratios studied in this experiment, the quality of the images of the test objects did not vary significantly. The capsules of ⁶⁰Co and ¹⁹²Ir were discernable.

The above experiments were repeated with a 5/8-inch-thick Pb filter, which was placed in front of the viewing screen. There was a modest improvement in image quality when ⁶⁰Co was the source of the secondary radiation. The improvement in image quality was somewhat greater when ¹⁹²Ir was the source of secondary radiation.

In a separate experiment, the ratio of the incident beam to the secondary radiation source was reduced to 3. Digital signal processing could have improved the image quality, which was satisfactory.

The resulting quality of the images was dependent upon the viewing system used. A GdOS screen coupled with an Isocon camera provided the best images, for instance. When using the GdOS screen and Isocon camera, the x-ray dose rate used above was too low to successfully image Drum #2. When the imaging system was changed to a high-energy image intensifier and a Newvicon camera, metal objects in the sludge were marginally detectable. Digital signal processing resulted in a marginal improvement in image quality. The golf ball-size pieces of Teflon-like material and the small plastic bottles were not

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visible, and the water on top of the plastic bag containing the sludge could not be seen when the beam had to first penetrate the sludge. Only in areas where the sludge was not present could the water be imaged.

Brown and Mikesell drew several conclusions from this set of experiments that are expected to be of use in any RTR system used to interrogate RH TRU waste drums destined for emplacement at the WIPP. Image enhancement techniques improved the image quality by increasing contrast. Optimization of geometric factors, control of secondary radiation via a collimator, and preferential filtration were all effective in improving image quality. The GdOS screen and Isocon camera were satisfactory for viewing the contents of Drum #1 but not Drum #2, which contained sludge. The high-energy image intensifier coupled with the Newvicon camera was satisfactory for imaging Drum #2, but the viewing area was small. The authors also felt that waste drums lined with a quarter-inch of lead would allow successful interrogation by the Minac-6 system, although they did not specifically examine such a configuration. Based on the experiments conducted, this is a reasonable conclusion.

III.C. 9 MeV "Linatron 300A" Electron Linear Accelerator System Experiments

Drum #1 was interrogated using the Linatron 300A system operated at 6 MeV with an output of 2,400 R/min at 1 meter, an output significantly higher than that of the Minac-6 system, which is 300 R/min at 1 meter. The detector system was a Varian ER110 that utilized a GdOS screen and an SIT (Silicon Intensifier Target) camera. A slight image enhancement was used. The resulting images were sharper than those obtained using the Minac-6 system.

When the ⁶⁰Co source was placed in the drum, the resulting images were not adversely affected by the added radioactivity. Experimenters were able to see the spray can, the bottles, and the liquid levels. Frame averaging provided some improvement in image quality without losing the ability to see movement of the liquid surface. Even when the ⁶⁰Co source was placed on the outside of the drum between the drum and screen, the capsule was easily visible with no apparent flaring or image degradation.

Drum #2 was interrogated with the Linatron 300A system operated at 9 MeV. The output of the system was 4000R/min at 1 meter. Experimenters evaluated several enhancement techniques, such as frame averaging, integrations of one-half to 8 seconds, and image subtraction, and determined optimal values for each technique.

The ⁶⁰Co capsule was inserted into the drum but was not visible unless contrast stretching and frame averaging were employed. Integration and image subtraction identified the location and approximate size of the ⁶⁰Co source. Aside from the threaded steel rod and the centerline pipe, there was no mention of imaging other objects within the drum.

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The ⁶⁰Co source was placed outside the drum and within 1½ inches of the screen. Exposure was estimated at 30,000 R/hour at the screen surface. When the experiment was repeated, there were no adverse effects observed.

In another experiment, ASTM (American Society for Testing Materials) steel penetrameters were taped to the outside of the drum nearest the source. These penetrameters ranged in size from 0.055 inches to 0.16 inches. The ⁶⁰Co source was placed on the outside of the drum nearest to the screen and opposite the center of the screen. The penetrameters were imaged and image enhancement techniques were applied. The outline of the 0.060-inch-thick 60 ASTM penetrameter was barely visible, and all three holes of the 0.140–inch-thick 140 ASTM penetrameter were visible. In this experiment, the outlines of most of the penetrameters were visible through 23 inches of sludge. In practice, as waste drums are rotated on their centerlines, the images would have to pass through only 12 inches of sludge; thus, it is reasonable to expect the resulting images to be sharper than those obtained in this test.

The authors concluded that the GdOS screen and SIT camera detector were quite satisfactory during the examination of both Drums #1 and #2. The Linatron 3000A system offered increased sensitivity over the Minac-6 system. The contents of waste drums lined with a quarter-inch of lead would be viewable using the Linatron 3000A system. Again, this appears to be a reasonable conclusion, although there was no specific experiment conducted to demonstrate this capability.

III.D Additional Reports and Documentation

A report entitled *Update of NDA and NDE Characterization Capabilities Since the Release of Remote Handled Transuranic System Assessment* (DOE/CAO-95-1143, LA-UR-00-6058),⁵ authored by Daniel P. Taggart of Los Alamos National Laboratory and submitted to the Carlsbad Field Office (CBFO) in December 2000, discusses the capabilities of non-destructive assay (NDA) and non-destructive examination (NDE) systems for characterizing RH TRU waste. This report states there is evidence suggesting that conventional RTR systems are capable of imaging the contents of TRU waste drums with gamma surface dose rates of "thousands of rads/hour." However, no references or data are given to support this statement. Images from a Bio Imaging Research Waste Inspection Tomography system using x-rays generated by a 2 MeV linear accelerator are included in the report. These images are of a concrete solidified drum and a steel pipe overpack and clearly show the contents of the drums. An image of a truck engine block that showed the piston rods of the engine, also included in the report, was produced by xrays generated by a 9 MeV linear accelerator manufactured by Bio Imaging Research. The report did not indicate surface dose rates for the waste forms involved in either of the above tests, however, and no other data were presented indicating that these systems would give acceptable results in the presence of high radiation fields emanating from the target. This system reputedly can image lead-lined glove boxes,

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though no data to substantiate this were provided. Taggart also describes an imaging system manufactured by SAIC, the Vehicle and Cargo Inspection System, which uses a Cs-137 source and a linear diode array as a detector. This system can reputedly image lead-lined glove boxes.

While Taggart imparts confidence that these systems would be usable for RH TRU wastes, he states that the performance of these systems in the presence of radiation fields emitted by the RH TRU waste has not been determined. He also suggests that one may subtract an image taken with the beam *off* from an image taken with the beam *on* as one approach to overcome high-surface dose effects.

Another report, *Characterization of RH-TRU and Lead-Lined Drums Using X-ray Imaging Techniques, INEEL/EXT-2001-00625*, ⁶ written by Timothy J. Roney of the INEEL and published in May 2001, indicates that surface dose rates associated with the RH TRU waste drums and high material density of the lead-lined TRU waste drums pose challenges to RTR systems. Roney cites a loss in image contrast and a subsequent loss in image quality, conditions which reduce an operator's ability to correctly identify prohibited items in the waste drums.

Roney evaluated the effectiveness of two systems—a collimated scanning linear diode array detector and an image intensifier system that records RTR data in a PC-based video format—to compensate for the above-mentioned interferences. He concluded that either system would "likely" provide sufficient image quality for "adequate operator interpretation" for the majority of RH TRU waste drums; however, no data on any actual tests of RTR on waste drums with surface dose rates in the RH range were provided. Another possible solution Roney addressed involved increasing the distance between the waste and the detector, which would minimize the reduction in image quality due to radioactive emissions from the waste. Effective RTR examination of lead-lined drums might be accomplished by using a higher energy x-ray source than that required for unlined drums. To overcome problems associated with examining lead-lined drums, the report suggests using a highly collimated scanning linear array detector.

Roney concluded that there may be TRU waste drums that RTR methods cannot satisfactorily examine due to three major factors: the wide variation in surface dose rates (200 mrems per hour to 1000 rems per hour); variation in the waste materials; and uneven distribution of the waste and radioactive sources. He further concluded that the actual performance of RTR on RH TRU waste drums "can only be confirmed by trials with surrogate or real drums."

In 2002 the National Academies Press published *Characterization of Remote-Handled Transuranic Waste for the Waste Isolation Pilot Plant: Final Report*⁷ (2002) (<u>http://fermat.nap.edu/catalog/10492.html</u>) in which it was noted that VE and RTR cannot distinguish between corrosive and non-corrosive liquids, whereas perhaps AK could make that distinction. Thus, AK may be a better way to identify prohibited

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items in TRU waste containers than RTR and VE. In addition, the report noted a lack of evidence to support the effectiveness of RTR in visualizing the contents of TRU waste containers across the entire range of permitted dose rates in RH TRU Waste (200 mrems per hour to 1000 rems per hour). The report also raised the concern that high neutron fluxes might impair the ability of RTR to provide adequate images of the contents of waste containers.

The National Academies Press report pointed out that although RH TRU waste is expected to account for approximately 14% of the total Ci activity, it will only account for a small fraction of the volume of waste emplaced at the WIPP. The report maintained that approximately 95% of the RH TRU waste scheduled for disposal at the WIPP "has yet to be generated or needs to be processed, packaged, or repackaged." According to the report, this TRU waste will be characterized by means other than RTR.

At the 2006 Waste Management Symposium, DOE representatives gave a presentation regarding the development status of an NDE and NDA system for TRU waste packed in large boxes and crates.⁸ The handouts provided at that presentation address some of the problems associated with re-packaging and characterizing the contents of large boxes; and they include a discussion regarding the development of a large box NDE and NDA system, which would minimize the need to repackage waste and allow characterization that meets the WIPP WAC. The handout material also describes a commercial system scheduled to be installed at the Savannah River Site (SRS) that produces a 30-degree cone of 3-MeV x-rays and includes a container table that allows for rotation, translation, and a \pm 10-degree tilt of large boxes of TRU waste, features that enable the system to image "sloshing" liquids.

This NDE and NDA system, scheduled to begin operations in October 2006, appears to be designed for CH TRU waste. (The handout did not address RH TRU waste.) Based on information from the copy of the presentation handouts provided to PECOS, this system might be able to successfully image some RH TRU waste, though no firm conclusions can be drawn regarding the suitability of this system to image RH TRU waste.

The two RH TRU waste characterization program documents^{9,10} prepared by DOE and the final Decision Letter from EPA that approved those documents¹¹ recognize that issues related to the use of RTR for characterization of RH TRU waste. They require that a demonstration proving RTR effective for the RH TRU waste streams for which it would be used be performed by the generator site as a part of the approval of that part of the RH TRU waste characterization program. However, there is no specific requirement in the program documents stating that a site's RTR equipment must be pre-qualified to confirm that higher dosage rates related to RH TRU waste do not affect the Data Quality Objectives established for RTR. Further, the program documents do not specifically require that the "known test targets" have surface dose rates in the range of RH TRU waste. Also noted was the fact that the reporting

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requirements for the performance of RTR on RH TRU wastes do not include a requirement to document the type of equipment used, equipment settings, or specifics of the "known test targets."

IV. CONCLUSIONS

Based on the information reviewed as summarized above, we present the following conclusions:

IV.A. RTR Can Be a Valuable Tool

The preferential methods for identifying prohibited items in waste containers destined for disposal at the WIPP are VE or AK. This applies to RH TRU waste as well as to CH TRU waste. However, RTR can be a valuable tool in the identification of prohibited items in the small percentage (<5%) of RH TRU wastes for which other methods are not practical or are difficult to execute. This includes RH TRU waste with high surface dose rates and waste in lead-shielded containers.

IV.B. Technology Advances Show Promise for Improved Effectiveness of RTR

The experiments reviewed for this report indicate that a high-energy x-ray source coupled with appropriate shielding and/or beam collimation (when necessary) and image enhancement techniques will provide an image of RH TRU waste that would enable operators to identify prohibited items that may be in the containers. In the years since the opening of the WIPP repository, there have been significant advances in the field of RTR. The emergence of high-energy x-ray generators with energies of 9 MeV hold promise for the examination of lead-lined waste containers. In addition, implementation of shielding and collimators placed in front of the detector as well as the practice of increasing the distance between the waste drum and the detector potentially offer improved image contrast and consequently, image quality when using RTR to image TRU waste containers with high radiation fluxes. Detector systems such as the linear diode array detector have also been applied to solving problems associated with imaging RH TRU waste containers. In essence, the progress being made in RTR interrogation systems and image enhancement techniques is very encouraging.

IV.C. Further Research Recommended by Previous Researchers Has Not Been Performed.

Taggart⁵ states that the technologies he mentioned in his report "would require further development before they could be used for RH-waste characterization." He further states that it should be demonstrated that an RTR system is capable of obtaining satisfactory images when the waste itself is highly radioactive. Taggart feels such a test would succeed, but that it "should be demonstrated conclusively as soon as

possible." Roney⁶ points out that due to the "high degree of variation in drum contents, matrix densities, matrix distributions, source intensity, source distribution," and other factors, the results he obtained should be considered preliminary. He suggests additional tests be conducted.

IV.D. RTR Not Fully Proven for RH TRU Waste Containers.

The RH TRU waste types that may be subjected to RTR prior to emplacement at the WIPP include but are not limited to: non-homogeneously distributed radioactive waste; waste with a high neutron flux; wastes contained in boxes or containers that cannot be transported in the existing shipping containers; and lead-shielded containers. Experiments to-date have been conducted on drums containing a single radioactive source, not on homogeneously distributed radioactivity. Thus far, no data have been presented to suggest that RTR has been tested under representative actual conditions for any RH TRU waste forms or types. The technique has not been proven effective on all unshielded RH TRU waste containers. Furthermore, few tests of RTR have been performed on RH TRU waste in concrete-filled or lead-lined containers, even though new instrumentation and techniques have been developed and tested for those types of containers.

V. RECOMMENDATIONS

The new RTR instrumentation and image enhancement techniques discussed in this report offer great promise and have a high probability of successfully imaging RH TRU waste that has high radiation fields and/or that is shielded with lead. It is reasonable to expect that prior to the time when a technique for the imaging of RH TRU waste is required, further advances in RTR instrumentation will be available. Nevertheless, good scientific practices dictate that before RTR is used to examine RH TRU waste containers, additional tests be conducted. One objective of further tests and experiments would be to demonstrate the acceptability of RTR in meeting all regulatory requirements related to sending RH TRU waste to the WIPP. A second objective would be to identify any limitations in the method before any RH TRU waste that has been characterized solely by RTR (even if this does comprise only a small percentage of the total waste stream) is shipped to the WIPP. Finally, a successful demonstration of an RTR system to examine the very small percentage of RH TRU waste that is not to be inspected by VE will show workers and public that the DOE is protecting their interests *and* the environment.

Therefore, it is recommended that:

1) DOE revise the RH TRU Waste Characterization program documents to ensure they specify the objectives of an RTR system approval test and an experimental design and test results DOE

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would consider necessary in order to meet those objectives and all requirements of the WIPP. Such a program should demonstrate the effectiveness of RTR on actual or surrogate RH TRU waste containers and should reflect the range of container types, internal shielding, and surface dose rates expected to be disposed in the WIPP.

- 2) While the RH TRU waste program documents allow each generator site to develop its own program, it would be prudent for all sites that expect to dispose of RH TRU waste requiring RTR to work together to design and test a system that meets all regulatory requirements. In fact, considering the small number of RH TRU containers that will require RTR testing, consideration should be given to assigning the Contractor Certification Program the responsibility of designing, testing, and implementing an RTR program that meets the requirements for all RH TRU waste streams expected to require RTR for characterization across the complex.
- *3)* Results of such a generic RTR testing program should be made public and approved by the regulators before any RH TRU waste is characterized by RTR and subsequently shipped to WIPP.

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