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Considerations for an Active and Passive CT Scanner to Assay Nuclear Waste Drums

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Introduction

Nuclear (or radioactive) wastes are generated at many DOE laboratories, military facilities, fuel fabrication and enrichment plants, reactors, hospitals, and university research facilities. At all of these sites, wastes must be separated, packaged, categorized, and packed into some sort of container—usually 208-L (55-gal) drums—for shipment to waste-storage sites. Prior to shipment, the containers must be labeled, assayed, and certified; the assay value determines the ultimate disposition of the waste containers. An accurate nondestructive assay method would identify all the radioisotopes present and provide a quantitative measurement of their activity in the drum. In this way, waste containers could be routed in the most cost-effective manner and without having to reopen them.

Currently, the most common gamma-ray method used to assay nuclear waste drums is segmented gamma-ray scanning (SGS) spectrometer [1] that crudely measures *only* the amount of ^{235}U or ^{239}Pu present in the drum. This method uses a spatially-averaged, integrated, emitted gamma-ray-intensity value. The emitted intensity value is corrected by an assumed constant-attenuation value determined by a spatially-averaged, transmission (or active) measurement [2]. Unfortunately, this typically results in an inaccurate determination of the radioactive activities within a waste drum because this measurement technique is valid only for homogeneous-attenuation or known drum matrices [3]. However, since homogeneous-attenuation matrices are not common and may be unknown, other NDA techniques based on active and passive CT* (A&PCT) are under development [4,5]. The active measurement (ACT) yields a better attenuation matrix for the drum, while the passive measurement (PCT) more accurately determines the identity of the radioisotopes present and their activities. We propose to use improved photopeak discrimination, simultaneous multiple-energy data acquisition, scatter rejection, and an ideal gamma-ray source to further improve the waste-canister analysis. Thus, no complicated corrections are required of the ACT data, in contrast to the approach of Kawasaki et al. [4], before it is applied to correct the PCT data. Some of our preliminary A&PCT results are given below.

Theory

Consider a single x - y plane fixed along the longitudinal axis, z , in a waste drum. We can treat a single discrete x - or gamma-ray beam in that plane as a line or ray path defined by s , the distance between the ray path and the $(x$ - y) origin and θ , the angle of this perpendicular from the x -axis. The transmitted-beam intensity $I(E,s,\theta)$ for this ray path at a fixed energy E is

$$I(E,s,\theta) = I_0(E,s,\theta) \exp \left[- \iint \mu(E,x,y) \delta(x \cos \theta + y \sin \theta - s) dx dy \right], \quad (1)$$

where $\mu(E,x,y)$ is the spatial distribution of the linear attenuation coefficients at energy E , I_0 is the intensity of the incident beam, and the equation for the ray path is $x \cos \theta + y \sin \theta - s = 0$. The argument to the exponential is known as a ray sum, $g(E,s,\theta)$, and is equal to

* In the text, we refer to emission CT as passive CT and transmission CT as active CT, since the second term in each pair is more common to the currently used nondestructive waste-analysis or waste-assay nomenclature.

$$g(E, s, \theta) = \ln \left[\frac{I_0(E, s, \theta)}{I(E, s, \theta)} \right] = \iint \mu(E, x, y) \delta(x \cos \theta + y \sin \theta - s) dx dy . \quad (2)$$

The set of ray sums at all values of s for a fixed E and θ is called a "projection," and a complete set of projections at all θ (over 180°) for a fixed E is called a "sinogram." From measurements of I and I_0 , a complete sinogram can be determined, and various methods have been devised to reconstruct μ , with filtered backprojection (FBP) being the most common [6]. Therefore, μ is the parameter determined by the ACT measurements.

Note that ACT does *not* yield the identity of the radioisotope (determined by the characteristic energy peaks emitted) or its activity (the intensity of its energy peaks) within a waste canister. On the other hand, PCT can be used to measure both. The ray sum for passive or single-emitted-photon CT (sometimes called SPECT) imaging, $g_\gamma(E, s, \theta)$, is defined by the integral equation

$$g_\gamma(E, s, \theta) = \iint p(E, x, y) a(E, x, y, s, \theta) \delta(x \cos \theta + y \sin \theta - s) dx dy , \quad (3)$$

where

$$a(E, x, y, s, \theta) = \exp \left[- \int \int_y^{\text{detector}} \mu(E, x', y') \delta(x' \cos \theta + y' \sin \theta - s) dx' dy' \right] \quad (4)$$

is the half-line attenuation integral from the (x, y) position to the detector position, defined by (s, θ) , and $p(E, x, y)$ is the activity measured in curies (3.7×10^{10} dps) of the passive source. Therefore, a single-emitted-photon ray sum is the summation of radioisotope activity, modified by an exponential attenuation, along the path to the detector. The influence of the term $a(E, x, y, s, \theta)$ depends on the magnitude and distribution of the attenuation coefficients within the waste drum, which unfortunately are quite large and nonhomogeneous for most energies emitted. For accurate PCT activity measurements, attenuation coefficients must be determined by ACT because the commonly used assumption of a constant attenuation coefficient is inadequate. For the work presented, we applied a steepest-descent-weighted least-squares technique [7] to reconstruct PCT images using attenuation correction from the ACT image according to Eq. (3).

Experiments

To demonstrate the quantitative possibilities of (1) ACT to obtain an x- or gamma-ray-attenuation image of a waste-canister interior; (2) PCT to locate and determine the radioisotopic sources present; and (3) coupled A&PCT to more accurately measure the radioactive isotope's activities, we used these two CT techniques to analyze a simulated waste canister. This 18-cm-tall, 11-cm-O.D. canister (~1/6 scale to a 208-L or 55-gal drum) resembles a sealed juice can and contains several items of diverse materials to simulate a heterogeneous-attenuation matrix. The canister contains several attenuating blocks (12.75×25.4 mm in cross section and with various lengths): one block each of copper, aluminum, glass, Lucite, and concrete. This canister was designed to allow access to the interior and to enable the interchange of various radioactive isotopes and their activities at different locations within the canister. For the A&PCT study discussed in this paper, a $95.5\text{-}\mu\text{Ci}$ ^{133}Ba disk-shaped (~4-mm-radius determined by autoradiography) source was inserted parallel to the glass block. ^{133}Ba has several energy peaks of interest and was chosen because it has a few energy peaks near those of the active source used, ^{192}Ir [8].

A medium-energy CAT scanner (MECAT) was used to acquire both the passive and the active CT data and is described in detail elsewhere [9]. This scanner uses a well-collimated source and detector

with first-generation scan geometry. The detector system consists of a high-purity intrinsic germanium detector and nuclear-spectroscopy instrumentation. This detector provides excellent energy resolution for (1) energy-peak discrimination at many peaks simultaneously and (2) scatter rejection by removing background counts. MECAT was slightly modified for this A&PCT study. The adjustable tungsten collimators were replaced by lead-brick ($5.0 \times 10 \times 20$ -cm) collimators with various fixed apertures.

For the PCT measurements, a 12-mm-aperture lead-brick collimator was placed at the front of the detector. Two PCT measurements were obtained. The first scan was centered ~118 mm from the canister bottom, and the net counts from the 81.0-, 276-, 303-, 356- and 384-keV peaks of the ^{133}Ba passive source were acquired simultaneously. The sinogram data set consisted of 45 projections at 8° intervals over 360° and 27 ray sums with a 6-mm translational step size. Each ray was acquired for 240 s to obtain a maximum of 24,088 emitted-net counts for a single 303-keV ray path. The second was acquired at approximately the same location, but for the source and its supporting box only—that is, without the simulated canister or its attenuating blocks. Net counts were obtained for all the above energies except 81 keV. The sinogram data-acquisition parameters were identical to those of the first PCT scan and resulted in a maximum of 35,598 emitted net counts for a single 303-keV ray path.

For the ACT measurements, a 6-mm-aperture lead-brick collimator was placed at the front of the source, and the detector collimator was the same as that used in the PCT scans. When this configuration was used, the count rate with a ~382-mCi ^{192}Ir source was greater than that allowable by the nuclear-spectroscopy-detector system. Because this was the only source available for performing the ACT scan, the count rate was decreased at the detector by moving the source to reduce the count rate. This resulted in a beam ~1.25 mm high and 5 mm wide positioned at the bottom of the detector aperture. The ACT slice plane was centered ~113 mm from the canister bottom. This is 5 mm below the center of the PCT scans. However, we do not expect this to dramatically affect the attenuation results because the blocks are invariant in z over this distance. Net counts from the 295-, 308-, 317-, and 468-keV peaks for the active ^{192}Ir source and the 303-keV peak of the ^{133}Ba passive source were acquired simultaneously. The sinogram data set consisted of 90 projections at 2° intervals over 180° and 54 ray sums with a 3-mm translational step size. Each ray was acquired for 30 seconds to obtain ~1.5% net-counting statistical uncertainty in the 295- and 302-keV energy peaks.

Results and Discussion

The ACT scans produce linear-attenuation-coefficient distributions in which the five blocks are easily distinguished (see images in Fig. 1). Clockwise from the top, these materials are Lucite, glass, copper, concrete, and aluminum. Their attenuation values agree with predicted values for the diverse blocks within experimental error. Also visible in the images are the canister itself, the ^{133}Ba source parallel to the long face of the glass block, and its internal supporting box. Notice that, as expected, the attenuation values decrease with energy.

We will describe the PCT results only for the 303-keV energy peak, as shown in Fig. 2. The PCT sinogram at 303-keV (Fig. 2a) reveals the areas where internal attenuation reduces the counts received at the detector. This sinogram was reconstructed without attenuation correction (see Fig. 2c). The position of the ^{133}Ba source is located where expected, relative to the ACT images. The reconstructed size of the source is large (~ 18×18 mm or 3×3 pixels) because the aperture used was 3 times the size of the source. Also, the source activity is approximately 2400 counts per pixel (attenuated), which is lower than

the measured activity (~4400 counts per pixel) obtained from the second PCT scan (unattenuated). To correct the PCT data for heterogeneous attenuation, the ACT images at energies 295 and 308 keV were linearly combined to generate an approximate attenuation image at 303 keV (see Fig. 2b). Correction by this image yields the attenuation-corrected PCT image (see Fig. 2d). Notice that the attenuation shadows of Fig. 2c are reduced in the corrected image, as expected. The activity values given in Fig. 2d are much higher than expected, and we are investigating the cause of this discrepancy, which we believe arises from the PCT reconstruction codes used.

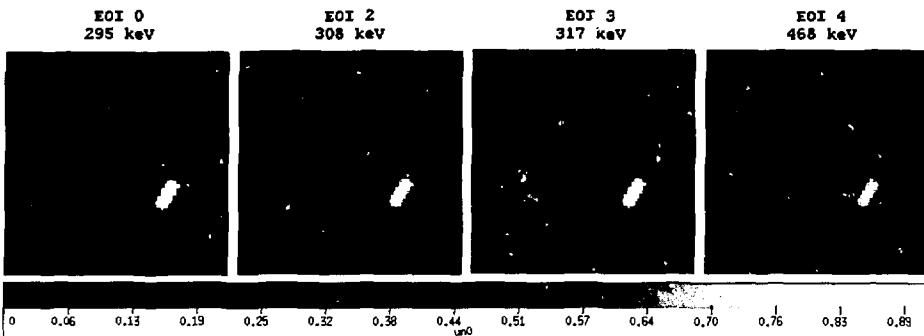


Fig. 1 ACT attenuation images reconstructed from 295-, 308-, 317-, and 468-keV sinograms for the simulated waste canister. All are displayed with the same gray scale given in units of cm^{-1} .

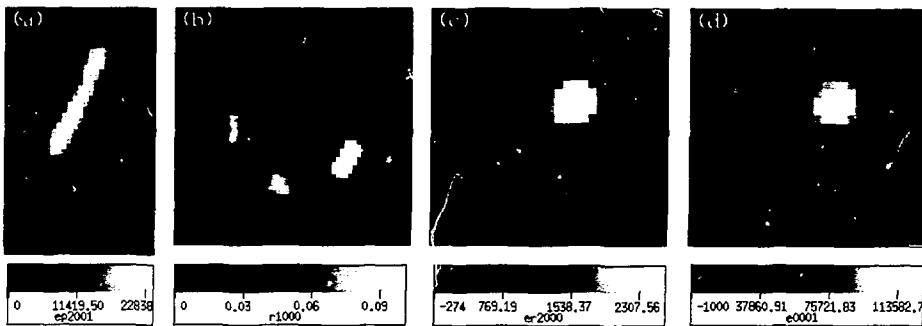


Fig. 2 PCT reconstruction results: (a) shows a representative PCT sinogram at 303-keV for the simulated canister and ^{133}Ba passive source; (b) is a weighted 303-keV ACT image determined from 295- and 308-keV reconstructions; (c) shows an image reconstructed from (a) with no attenuation correction; and (d) shows an image reconstructed from (a) using the attenuation distribution from (b). The gray scales for (a), (c), and (d) are in units of photon counts per 240 s, and (b) is in units of cm^{-1} .

Conclusions

Active and passive CT measurements using energy-dispersive detection systems can vastly improve the nondestructive assay of radioactive waste containers. Preliminary results show that the attenuation correction at individual energy regions-of-interest can result in quantitative image reconstruction. Currently, work is in progress to determine the discrepancy in the PCT corrected images and to better understand the relations among the four performance parameters: space, contrast, energy, and temporal (or speed) resolution from the point of view of the radioactive assay of nuclear wastes. Physical limits to improving the PCT parameters result from blurring caused by the collimator's angular cone of acceptance, photon scattering and starvation, the randomness inherent in photon counting, poor detection efficiency, the single-photon counting required for energy discrimination, and system noise. Quantitative assay using PCT is further complicated by the need for attenuation correction and scatter compensation.

There are two main challenges for the design and development of a prototype A&PCT scanner. The first is to incorporate the important results obtained from the coupling of energy-specific active and passive CT measurements, plus results gained from measurements to determine minimum detectable limits. The second challenge is to see if a CT scanner can be designed that will optimize both active and passive CT data acquisition and analysis to quantitatively assay nuclear waste drums.

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