Assay of Uranium Oxide Product Canisters Using Constrained Tomographic Gamma Scanning (TGS)

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The TGS (Tomographic Gamma Scanner) method has been applied to the assay of ^{235}U in UO_2 product cans. Pure UO_2 is too dense to be assayed by the standard TGS assay using the 186-keV gamma ray because of very high self-shielding and the inability of normal transmission sources to penetrate the cans. Fortunately, there are variations of TGS that will do a good job for assaying these cans. These take advantage of prior knowledge of the sample to force TGS_FIT software images into known constraints. The approach described here forces the transmission and emission images into a fixed shape with layer densities corresponding to the average log-transmission of the transmission gamma rays through the UO_2 component of a tailored material basis set (MBS). The use of the MBS formalism allows the user to specify the container and UO_2 geometry with separate "partial density" images of the can and UO_2. The usual TGS attenuation corrections are applied to this constrained image to give the ^{235}U content. The only assumptions in the method are that (1) in a given sample, the isotopic and chemical composition of the UO_2 is uniform throughout and (2) that there are no significant voids of non-UO_2 filler that are not accounted for in the constraints. The method is demonstrated to give accurate ^{235}U assays in this difficult application.

1. INTRODUCTION

The mission of the PERLA Laboratory at the Joint Research Centre in Ispra, Italy, is to provide facilities for safeguards NDA measurements, training of personnel, and testing of NDA techniques. As part of this mission, the PERLA Laboratory is evaluating the use of the TGS (tomographic gamma scanner) method[1] for improved safeguards NDA of the ^{235}U content in product UO_2 cans, using an Antech Corporation can TGS system. Figure 1 shows the design of the product cans studied, which come in three sizes. All have the same 8.7 cm inner diameter and are made of stainless steel. The outer walls are 0.4 cm thick, with a 0.2 cm thick bottom and 0.6 cm thick top. The cans are designated by the model numbers ISPRA 2000 (1.7-L volume), ISPRA 1000 (0.84-L volume), and ISPRA 500 (0.49-L volume).

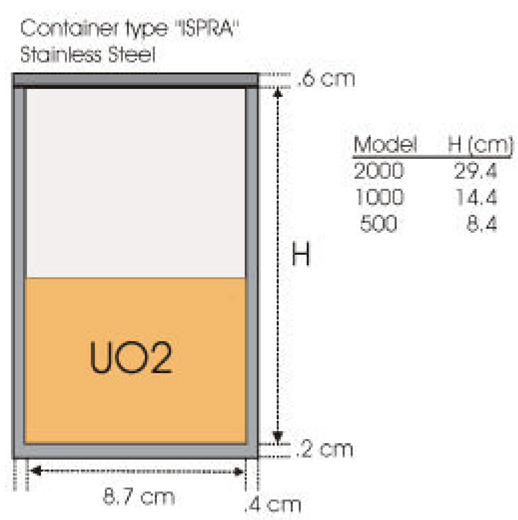

FIG. 1. "ISPRA" cans for storage of UO_2. The cans come in three sizes of varying height and volume, but with the same diameters. The approximate maximum volumes of UO_2 that the cans will hold are 1.7-L (model 2000), 0.84-L (model 1000), and 0.49-L (model 500). The UO_2 fill height is variable.

Because of self-shielding in the UO_2, assay of the Ispra product cans is a significantly more difficult problem than assaying the diffusely distributed uranium found in typical waste. The UO_2 is undiluted by matrix materials in the product cans, and it fills the containers more or less uniformly to some effective fill height. UO_2 has a linear gamma-ray attenuation coefficient of μ = 14.259 cm^{-1} for the 186-keV gamma ray from ^{235}U. Thus, the 186-keV gamma rays emitted come from a mean depth of only 0.07 cm, with 99% coming from the outer

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0.32 cm. Thus, only a thin outer shell can be assayed by gamma emission methods. Neutron assays are at least as problematic in this application as are gamma assays. Self-shielding of the same magnitude exists with the DDA technique (DDT). In that case, the interrogating thermalized neutrons can only penetrate a thin outer shell, so again only the outer skin of the sample can be assayed directly. Passive neutron counting cannot be used because of the low spontaneous neutron emission rate of the uranium isotopes.

TGS transmission imaging is also difficult with the UO_2 product cans. The 401-keV gamma ray, the highest energy gamma emitted from the ^{75}Se sources normally used for TGS transmission imaging, cannot penetrate the UO_2 except near the edges of the can. Its transmission at the thickest part of the sample is less than 10^{-11}. A transmission of approx. 0.003 can be achieved at the thickest part of the can using the 1,333-keV gamma ray of ^{60}Co. Thus, a strong ^{60}Co transmission source used in combination with ^{75}Se or ^{133}Ba could be used to make faithful density images of the UO_2 layers in addition to the low-density, empty part of the cans. However, while this might give accurate and repeatable transmission images, it still would not be possible to make a good TGS emission image of the ^{235}U gamma rays because almost no gammas can be seen except those from the outer 0.32 cm of the UO_2. Even an idealized emission imaging algorithm analyzing noise-free data could at best see only a shell of ^{235}U surrounding the cylindrical volume of UO_2, and would have a response that is proportional to the surface area. The actual algorithms used in standard TGS—expectation maximization (EM) and the algebraic reconstruction technique (ART)—tend to find at least some ^{235}U in the dense core of the sample as image clutter, which results in erratic assay results.

Fortunately, the Los Alamos TGS_FIT software,[2] which is used as the kernel for the Antech MasterAnalysis software, offers constrained variations on the normal TGS analysis that will do a good job on these cans. The key is to take advantage of prior knowledge of the sample to force the images into known constraints. The approach we used is to force the transmission and emission images into a fixed shape with layer densities corresponding to the average log-transmission of the transmission gamma rays through the UO_2 component of a tailored material basis set (MBS). The use of the MBS formalism allows us to specify the container and UO_2 geometry with separate "partial density" images of the can and UO_2. In this case, we model the container as being composed of a basis set of stainless steel (SS) and UO_2. The SS can density image is treated as fixed, while the UO_2 density image is constrained to be within the can at varying layer densities that are constant within each layer. This determines the spatial distribution of the UO_2, subtracting out the loss due to the fixed SS can. This technique gives excellent images even with the poorly penetrating ^{75}Se as a transmission source, because the transmission at the edge of the can is sufficient to predict the density of the UO_2 layers. In practical terms, this transmission image will usually correspond to the effective fill height. However, it is important to recognize that this is the actual distribution as determined by the material's density, and not merely the fill height. In particular, if the surface of the powder is canted at an angle, this will show up in the transmission image as additional reduced density layers at the surface of the UO_2.

In the second part of the analysis, the UO_2 transmission image is transferred to the emission image space, where it is assumed to give the correct *relative* distribution of ^{235}U. The TGS attenuation correction is applied to this relative distribution to obtain the attenuation-corrected total response as the 186-keV gamma count rate per gram of ^{235}U. Comparing this to the measured total count rate gives the total ^{235}U mass in the can.

This procedure gives a true assay of the ^{235}U content, provided that the following assumptions are met: (1) the UO_2 is of uniform chemical and isotopic composition within any individual sample, and (2) there are no significant voids or regions occupied by other filler material in the UO_2 volume. There is no requirement that the density of the material be uniform from can to can or from layer to layer within a can, since we are measuring the density of each layer. If the product in some cans is fluffier than is usual or if the surface is canted, this will be accounted for in the density images. Moreover, the isotopic composition can vary from can to can. Measurement of the emission rates of the 186-keV gamma assures that the ^{235}U content is being measured, not the UO_2 content (although that comes out indirectly from the UO_2 density image).

2. EXPERIMENTAL DETAILS

Table 1 lists seven standard UO_2 cans of uranium oxide that were scanned for the evaluation. At least one of each of the three different can sizes was represented, with ^{235}U masses ranging from 24.87 to 348.2 g.

The scans were performed using an Antech can scanner running the Antech MasterScan software. Similar Antech can scanners are in use at Rocky Flats and at Los Alamos. Ordinary two-pass TGS scans were performed to collect the data. Table 2 lists the scanner settings used. Note that the scan extents (scan diameter and height) are significantly larger than the dimensions of the product cans. This is acceptable in TGS (the rule is that the scan extents must be equal to or greater than the size of the sample scanned).

3. TGS_FIT DATA ANALYSIS

Methods and commands for applying the TGS_FIT analysis and image reconstruction software to analyzing the Ispra can data are discussed in this section.

Additional details and discussions of the commands can be found in *The TGS_FIT User's Manual*.[2]

A. Tailoring the Material Basis Set

The MBS formalism is introduced in Estep, Prettyman, and Sheppard (1995) and is described in more detail in Estep, Prettyman, and Sheppard (1996) and later articles. We ordinarily use the data space coupling scheme in TGS analyses with a material basis set of (say) Z=5 and 82, or some similar Z points, to create a "computed" MBS. That is, TGS_FIT uses its built-in attenuation factor model to compute attenuation coefficients at all of the transmission and emission energies for fixed densities of Z=5 and 82. In that description, the canister and UO_2 true transmission images would be mingled together, although most of the UO_2 would be in the Z=82 rho-image (a rho-image is the MBS partial density vector for a material in the basis set). In this problem, we use full coupling as described by Eq. (3) in *User's Manual for TGS_FIT Version 2.0*,[2] and a material basis set of SS and UO_2. We compute the attenuation coefficients manually with the PHOTCOEF program from Oak Ridge. These go into the energy template (.ZMT) file, which specifies the emission and transmission energies and the material basis, as "TABLE" values instead of "COMPUTED." The ZMT file we used here, named U235CM.ZMT (the *User's Manual for TGS_FIT Version 2.0* contains a complete description of ZMT files[2]) is:

```
4 3 2
136 266 280 401 143 163 185
26 82
0 1 2 3 4 5 6
1 1 1 1 1 1 1
TABLE
1.8045 31.981
.94134 6.7290
.90805 6.0059
.73926 2.9350
1.6675 28.310
1.4198 20.672
1.2462 15.288
NONE
```

Note that though line 3 specifies Z=26 and 82, these are used only as placeholders. The MBS method is applied using the table values of the attenuation coefficients at the bottom, which are in units of 1/cm. The two columns under "TABLE" list the attenuation coefficients of SS and UO_2 at the transmission and emission energies listed on line two. To allow the use of attenuation coefficients in 1/cm, we built the response matrices (in TGS_MAT) using units of centimeters rather than inches.

To generate images of the can and the UO_2, we wrote a Visual Basic program called "Add-a-can.exe." This program creates TGS_FIT command (.PRO) files that implement the TGS_FIT **EDIT U** command, as shown in the partial listing of file CAN2000.PRO, which models or simulates the empty canister:

```
;Generated by Add-a-can.exe
CLEAR U
EDIT U
0 24 0.1849694
0 25 0.1849694
0 33 0.2965715
0 34 0.1229479
0 35 0.1229479
0 36 0.2965715
... (lines skipped)
0 66 0.2965715
0 74 0.1849694
0 75 0.1849694
1 24 0.1707406
1 25 0.1707406
1 33 0.2418826
1 34 2.845678E-02
1 35 2.845678E-02
... (more) ...
```

The image created has a diameter of 9.5 cm, a bottom of thickness 0.2 cm, a top of thickness 0.6 cm, and cylindrical walls of thickness 0.4 cm of iron. The

TBL. 1. UO_2 Standards Used.

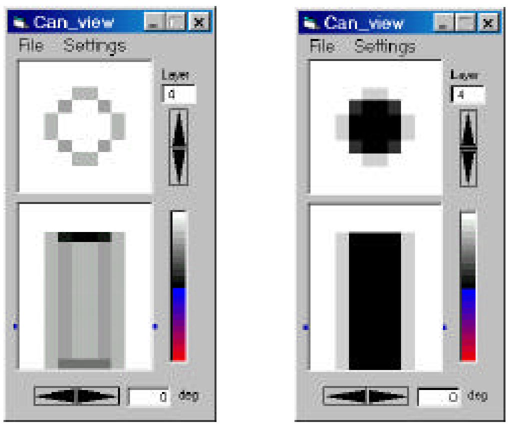
Identifier	Can Model	^{235}U mass	Number of Scans
U41	ISPRA 500	24.87 g	2
U42	ISPRA 500	49.74	2
U43	ISPRA 500	79.95	3
U44	ISPRA 1000	99.5	5
U48	ISPRA 2000	248.6	6
U49	ISPRA 2000	298.4	4
U50	ISPRA 2000	348.2 g	7

TBL. 2. TGS Scanner Settings.

Parameter	Value/Description
Scan height	31.75 cm
Scan diameter	21.59 cm
Number of layers	15
Views per layer	150
Voxels per layer	100
Collimator/detector type	Circular (both)
Collimator distance	13.97 cm
Collimator depth	7.62 cm
Detector diameter	5.715 cm
Layer coupling	+/-4

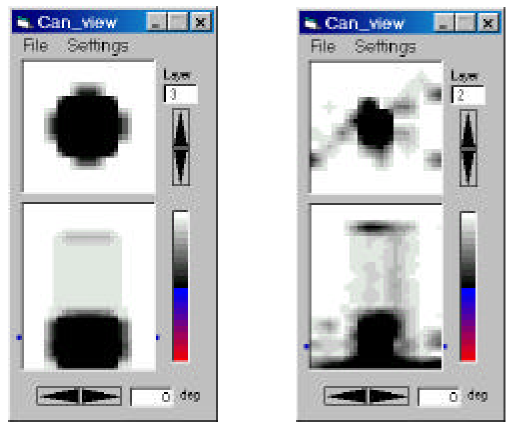
```
...
Transmission scan: layer 1, file= , peak=0
i= 0 :0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
i=10 :0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
i=20 :0.000 0.000 0.000 0.000 0.171 0.171 0.000 0.000 0.000 0.000
i=30 :0.000 0.000 0.000 0.242 0.028 0.028 0.242 0.000 0.000 0.000
i=40 :0.000 0.000 0.171 0.028 0.000 0.000 0.028 0.171 0.000 0.000
i=50 :0.000 0.000 0.171 0.028 0.000 0.000 0.028 0.171 0.000 0.000
i=60 :0.000 0.000 0.000 0.242 0.028 0.028 0.242 0.000 0.000 0.000
i=70 :0.000 0.000 0.000 0.000 0.171 0.171 0.000 0.000 0.000 0.000
i=80 :0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
i=90 :0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
...
```

FIG. 2. The CAN2000.PRO command file in TGS_FIT creates this image for a middle layer (as output by the **SHOW U** command).



(a) (b)

FIG. 3. Base images of the (a) can and (b) UO_2 generated by the ADD-A-CAN software, as viewed using the ANVIEW image viewing software. These are used as constraints in transmission imaging. The can image on the left corresponds to the **SHOW U** command listing in Fig. 2.



(a) (b)

FIG. 4. (a) Constrained and (b) unconstrained transmission images of standard can U49.

height depends on the can model. Running the CAN2000.PRO command file in TGS_FIT creates the image shown in Fig. 2 for a middle layer (as output by the **SHOW U** command):

The AN_VIEW display software image of the SS basis as shown in Fig. 3(a) is more revealing. Notice the thin bottom and thick top. The UO_2 component is modeled as a solid cylinder, as shown in Fig. 3(b). These are the dimensionless "rho-images," not ordinary u-images (attenuation coefficient images), in TGS_FIT. When projected to attenuation coefficient space, the two images would be blended in some proportion.

B. Fitting the Rho-Images by Scaling

We know that the UO_2 powder will fill the container from the bottom up, and that it will be approximately uniform up to its fill level (although possibly not level, a minor issue). Therefore, we require the UO_2 to be uniform within the canister, just as we generated it in the UT2000.PRO image file, except with variable layer densities. To fit the UO_2 transmission image within those constraints, we use the TGS_FIT **TRANS SCALE_RHO** command, which modifies the existing non-fixed rho images by scaling them layer by layer to give the best match to the log-transmission data (t-data) in the four transmission energy sets. In this case, only the UO_2 rho-image is scaled, while the SS rho-image (of the can) remains fixed. The SS canister contribution is essentially subtracted from the log-transmission data, and the UO_2 rho-image is scaled to match what is left. The result is an excellent UO_2 plus can image, as shown in Fig. 4(a) for the U49 standard (tall can), even when using the poorly penetrating ^{75}Se source. The “normal” unconstrained TGS image for the same data is shown in Fig. 4(b) for comparison.

Figure 4(a) is an interpolated u-image for the 186-keV emission line, and so combines the canister and the UO_2 images, which are still separate as rho-images.

Note the smearing out of the unconstrained TGS image in Fig. 4(b) at the bottom layer and in the cross sectional view. This occurs because of the poor penetration of the transmission source. With a more penetrating source, such as ^{60}Co, these artifacts should be significantly reduced. However, even then the unconstrained transmission image can never attain the accuracy of the constrained image in Fig. 4(a). To the extent that the constraints are valid, the constrained image in Fig. 4(a) represents the best use that can be made of the TGS transmission data.

C. Fitting the Emission Images by Scaling

To obtain an emission image of the 186-keV gamma ray, the UO_2 rho-image is copied directly to the emission image (s-image) vector for that energy, using the **COPY_U2S** and **COPY** commands. The assumption is that the UO_2 rho image, which gives the mass density of UO_2 relative to its standard state of 10.96 g/cm^3, is identical to the emission image except for a scaling factor. To scale the emission images, we use the **TRANS NORM_MBS** command. Unlike the **SCALE_RHO** command, the layers are not scaled independently, but all at once. An emission image obtained this way from an assay of standard number U49 is shown in Fig. 4.

4. RESULTS AND DISCUSSION

Figure 5 shows the results from assaying the seven standards listed in Tbl. 1, using ordinary TGS analysis and the constrained method described in Sec. 3. The improvement in the results with the constrained fitting is dramatic. Without the constraints, the results are erratic and the method would be considered unreliable. With the constraints, the results are accurate and the method can be recommended.

The use of a ^{60}Co transmission source would allow a more generalized approach in which the transmission image is less constrained. In this variation, the constraint on the reconstructed rho-image for UO_2 is that it be less than or equal to the starting (constraining) UO_2 image in every voxel. That is, the reconstructed UO_2 rho image would be constrained to be inside the can and to be no denser than UO_2 in the powdered state of 10.96 g/cm^3, but otherwise would be unconstrained. This variation would allow unusual shapes of UO_2, such as clumps or voids, to be handled more effectively.

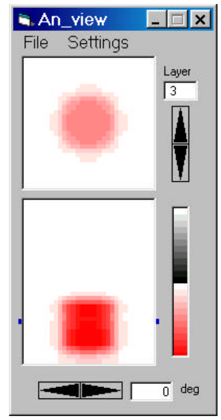


FIG. 5. A constrained emission image of 186-keV gamma rays from standard U49.

The success of using constrained images for the assay of product UO_2 suggests other potential applications. In particular, a similar assay could be performed on uranium or plutonium metal components, which are even more self-shielded than UO_2. Because the shapes encountered could be fairly complex, the more general method described in the previous paragraph would be required. It seems likely that as long as the transmission image of the metal is faithful and has sufficient detail, and the materials surrounding the metal object are known and can be compensated for, a fairly accurate estimate of an item's mass could be made in this way.

1. R. J. Estep, T. H. Prettyman, and G. A. Sheppard, "Tomographic Gamma Scanning to Assay Heterogeneous Radioactive Waste," Nucl. Sci. and Eng. **118** 145-152 (1994).
2. R. J. Estep, *User's Manual for TGS_FIT Version 2.0*, NIS6-QAP-00.33, Rev 2.0 (Los Alamos, 2000).
3. R. J. Estep, T. H. Prettyman, and G. A. Sheppard, "Reduction of TGS Image Reconstruction Times Using Separable Attenuation Coefficient Models," *ANS Winter Meeting Transactions, San Francisco, California, November 1995.*
4. R. J. Estep, T. H. Prettyman, and G. A. Sheppard, "Comparison of Attenuation Correction Methods for TGS and SGS: Do We Really Need Selenium-75?," *Proc. 37th Annual INMM Meeting, Naples, Florida, July 28–31, 1996.*.