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AUTHOR(S) G. A. Sheppard, P. A. Russo, T. R. Wenz, M. C. Miller,
E. C. Piquette, F. X. Haas, J. B. Glick, and A. G. Garrett

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Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

MODELS FOR GAMMA-RAY HOLDUP MEASUREMENTS AT DUCT CONTACT[*]

G. A. Sheppard, P. A. Russo, T. R. Wenz,
M. C. Miller, and E. C. Piquette
Los Alamos National Laboratory
Los Alamos, New Mexico 87545 USA

F. X. Haas, J. B. Glick,
and A. G. Garrett
EG&G Rocky Flats, Inc.
Golden, Colorado 80401 USA

ABSTRACT

The use of gamma-ray measurements to nondestructively assay special nuclear material holdup in DOE processing facilities has increased recently. A measurement approach that is relatively insensitive to deposit geometry involves withdrawing the detector from the holdup-bearing equipment far enough to validate an assumed point-, line-, or area-source deposit geometry. Because of facility constraints, these generalized geometry procedures are not always followed, and some ducts are measured at contact. Quantitative interpretation of contact measurements requires knowledge of the width of the deposit transverse to the duct axis. Rocky Flats personnel have introduced a method to obtain data from which this width can be deduced. It involves taking measurements in pairs, with the detector viewing the holdup deposit at contact from above and below the duct. The interpretation of the top and bottom measurements to give the deposit width at each location requires a model for the detector's response to radial source position and a model for the deposit geometry. We have derived a relationship between the top-to-bottom count rate ratio and the deposit width that approximates the detector response and models the deposit geometry as a uniform strip. The model was validated in controlled experiments that used thin foils of high-enriched uranium metal to simulate duct deposits.

INTRODUCTION

The use of gamma-ray measurements to nondestructively assay special nuclear material holdup in the exhaust ductwork in DOE processing facilities has increased recently. Generalized (point, line, or area) deposit geometries within the detector's cylindrically collimated field of view are recommended for holdup assays.[1-3] The generalized geometry approach requires withdrawing the detector from the holdup-bearing equipment far enough to validate the assumed point-, line-, or area-source geometry. Assays of point, line, and area deposits viewed by cylindrically collimated detectors are based on calibrations obtained with point reference sources of the special nuclear material. In the calibration procedure, the detector response is the weighting factor that multiplies the incremental length or area in the sum that is used to compute the "effective width" or "effective area," respectively, for the field viewed by the detector. These effective geometric parameters are used with the measured count rate for the point reference source centered in the detector field of view to give the assay calibrations for line and area sources. The assay results corresponding to the point, line, and area deposit geometries are given in mass, mass per unit length, and mass per unit area, respectively. For the line and area deposits, the assay results must be multiplied by a deposit length and area, respectively, to obtain the net holdup assay. This analysis assumes that the deposits lie primarily along the bottom surfaces of the ducts and that the deposit thicknesses are uniform and thin enough that the effects of gamma-ray self-attenuation can be neglected.

Although many aspects of the generalized geometry methodology are used in holdup measurements within the DOE complex, other procedures are not always performed because of facility constraints. Certain procedures established at the Rocky Flats Plant, for instance, required that the ducts be measured at contact. The ducts at Rocky Flats vary in diameter from 4 to 15 in. Piping and other plutonium-bearing process fixtures are closely spaced or interwoven with the ductwork. The advantages of measuring the ducts at contact using well-collimated detectors are improved signal-to-background ratios and fixed detector-to-duct (but not necessarily detector-to-source) geometries. However, contact measurements suffer large uncertainties from local variations in the mass of the deposit. Therefore, a larger number of contact measurements is required to achieve the sampling equivalent to that obtained with a wider field of view at a greater distance. Furthermore, quantitative interpretation of contact measurements requires knowledge of the width of the deposit transverse to the duct axis. This is because the detector is not sufficiently distant to neglect the deposit width and validate the line source assumption, as in the generalized geometry approach.

Rocky Flats personnel have introduced a method[4] to obtain the width information required for quantitative interpretation of contact measurement data. The technique involves taking measurements in pairs, with the detector viewing the holdup deposit at contact from above and below the duct. The detector collimator was designed so that, at a distance of one duct diameter, the diameter of the detector's field of view was slightly larger than the diameter of the duct. Figures 1(a) and 1(b) illustrate the actual geometry of the detectors relative to the duct and its deposit. The deposit is depicted as a continuous ribbon along the bottom of the duct.

Because the duct diameter is significantly larger than the collimator apurture, the deposit geometry for a contact measurement from the bottom of the duct is best described as an "area" source. The holdup mass per unit area (areal density) for this measurement is obtained by multiplying the bottom count rate by the area source calibration constant. If the deposit is thin enough that self-absorption is negligible, this value is a good estimate of the areal holdup density within the detector's limited field of view. Unless there is confidence that the deposit is uniform along the duct, the small size of the sampled area requires a large number of samples to adequately characterize the entire deposit.

When the detector is positioned on top of the duct looking down, it views a larger, and therefore more representative, deposit sample. However, the deposit from this view is unlikely to fill the detector field of view. Application of the generalized-area-geometry assumption leads to an underestimate of the deposit areal density in this case.

DETERMINATION OF DEPOSIT WIDTH

The interpretation of the top and bottom measurements to give the deposit width at each location requires a model for

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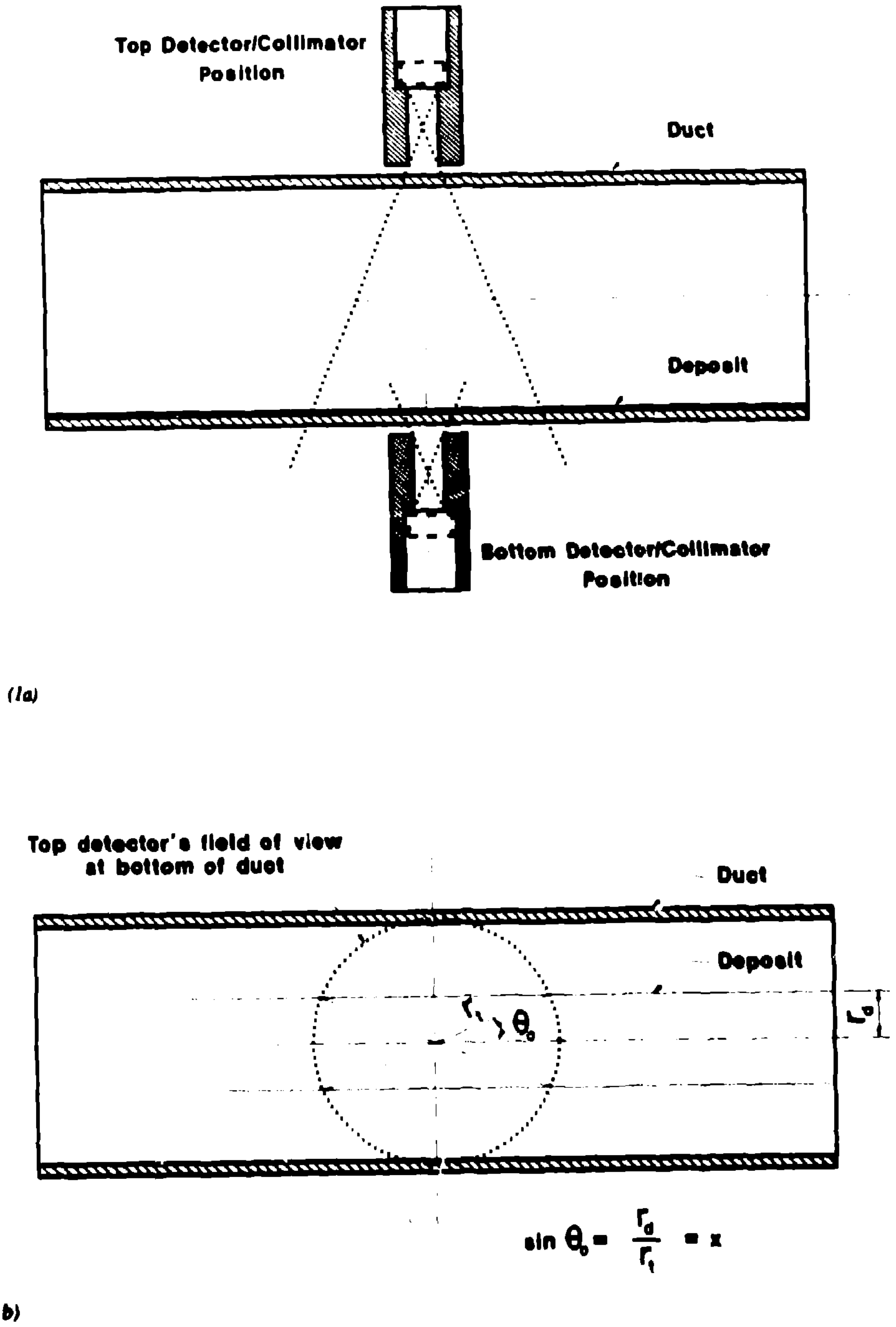

Fig. 1(a). Side view of detector/duct/deposit geometry. (b) Top view of detector/duct/deposit geometry. Diameter of top detector's field of view is slightly larger than the duct's outside diameter. Diameter of bottom detector's field of view is assumed to be smaller than deposit width.

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the detector's response to radial source position and a model for the deposit geometry. Once a deposit width has been deduced, it can be multiplied by the area-source assay result to yield a mass per unit length at that duct location. Then the holdup mass between each pair of adjacent locations is computed by multiplying the distance between them by the average of their mass-per-unit-length assay results. The total isotope mass in the duct is obtained by summing the holdup masses between all adjacent pairs of measurement locations.

Rocky Flats Approach

Rocky Flats personnel have interpreted their top and bottom contact measurements by assuming the following:

(1) The deposit is on the bottom half of the inside surface of the duct.

(2) The detector field of view is filled by the deposit for the measurement from the bottom.

(3) The detector field of view may not be filled by the deposit for the measurement from the top.

(4) Viewed from the top, the deposit is a uniform disk concentric with the detector field of view.

(5) For a given distance from the detector, the detector response is constant for all radial locations within the detector field of view.

With these assumptions, it is simple to derive the relationship between the deposit half-width, r_d, the radius, r_t, of the top detector's field of view at the bottom of the duct, and the measured net count rates, C_B and C_T, for the bottom and top measurements, respectively:

$$\frac{r_d}{r_t} = \left(\frac{C_T}{C_B}\right)^{1/2} \tag{1}$$

This relationship was used in the Rocky Flats method to determine the deposit width, $2r_d$. This width was multiplied by the area-source assay result (isotope mass per unit area) determined from the top count rate to obtain the isotope mass per unit length at the measurement location.

Alternative Approach

Given that the detector/collimator dimensions are small compared to the duct diameters and that the diameters of the fields of view for the top measurements are slightly larger than the duct diameters, assumptions (2) and (3) in the above approach are reasonable. However, realistic deposits should tend to be continuous along the length of a duct rather than appearing as disk-shaped spots of variable diameter concentric with the detector field of view. Furthermore, the assumption of a constant detector response across the detector field of view departs significantly from the true response as indicated in Fig. 2. The following simple alternatives to assumptions (4) and (5) are proposed:

(4') The deposit is a uniform strip running along the duct's length through the center of the detector field of view. The deposit length spans the detector field of view and its width is less than the duct diameter.

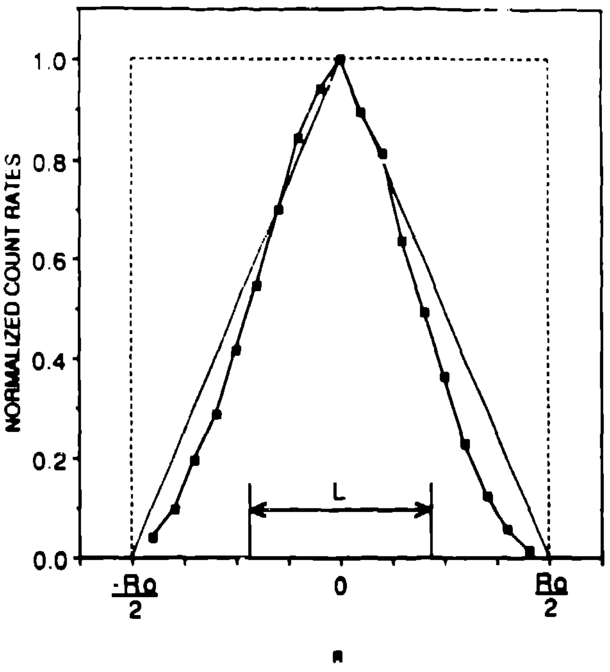

Fig. 2. Response of cylindrically collimated sodium iodide detector (2.54-cm diameter and 1.27 cm thick) to the radial position of a highly enriched uranium point source. Collimator is 1.90 cm in diameter and 3.81 cm deep and R_0 = 40 cm. Solid lines and squares indicate measured response. The dashed curve is the flat detector response assumed in the original Rocky Flats width model. The dotted curve is the triangular response assumed by the new Los Alamos width model.

(5') The detector response is triangular, with unit response at $r = 0$ and zero response at $r = r_t$. Thus, the normalized response function for positive values of r is

$$I(r) = 1 - \frac{r}{r_t} \tag{2}$$

Assumptions (1), (2), (3), (4'), and (5') can be used to obtain an improved expression that relates the measured count rates, C_B and C_T, to the deposit half-width, r_d. First, determine A_T, the "effective" deposit area viewed by the top detector, by integrating over the deposit surface weighted by the detector response:

$$A_T = 4\int_0^{\theta_0}\int_0^{r_t}\left(1 - \frac{r}{r_t}\right) r\,dr\,d\theta + 4\int_0^{\theta_0}\int_0^{\frac{r_d}{\sin\theta}}\left(1 - \frac{r}{r_t}\right) r\,dr\,d\theta$$

$$= \frac{2}{3} r_t^2 \left\{\sin^{-1} x + 2x\left(1 - x^2\right)^{1/2} + x^3 \ln\left[\frac{1 - \left(1 - x^2\right)^{1/2}}{x}\right]\right\}$$

where $x = r_d/r_t$. Second, determine A_B in a similar manner for the bottom detector:

$$A_B = 2\pi \int_0^{r_i} \left(1 - \frac{r}{r_i}\right) r\, dr = \frac{\pi}{3} r_i^2$$

The ratio A_T/A_B is equal to the count-rate ratio, C_T/C_B.

$$\frac{A_T}{A_B} = \frac{C_T}{C_B} = \frac{2}{\pi}\left\{ \sin^{-1}x + 2x\left(1 - x^2\right)^{1/2} + x^3 \ln\left[\frac{1 - \left(1 - x^2\right)^{1/2}}{x}\right]\right\} \tag{3}$$

The results of this model are plotted in Fig. 3, along with the results of the Rocky Flats model. The model predictions agree when $C_T/C_B = 1$. This is the case when the deposit width viewed from the top is equal to the diameter of the detector field of view. As the count rate ratio decreases, however, a large deviation is observed between the two models, with the Rocky Flats model giving the larger deposit width for a given top-to-bottom count rate ratio.

TESTING THE WIDTH MODELS

Preparation of Measurement Standards

Holdup deposits were mocked up using a set of uranium foils enriched to 93.15% in ^{235}U. The foils, nominally 46 cm long and either 3.8 or 7.6 cm wide, range in thickness from 0.027 g/cm^2 to 0.056 g/cm^2 (see Table I). They were laminated between sheets of 0.008-cm-thick plastic to protect them and to control contamination. The foil thicknesses were characterized by measuring the transmitted intensity of a well-collimated beam of 122-keV gamma rays from a ^{57}Co source. Transmission measurements on the 7.6-cm-wide foils were made at 216 equally spaced locations. The 3.8-cm-wide foils were measured in 108 places. Table I includes the mean thickness and standard deviation for each foil, along with the effective ^{235}U thickness. The latter is the mean thickness multiplied by the enrichment and corrected for self-attenuation at 186 keV.

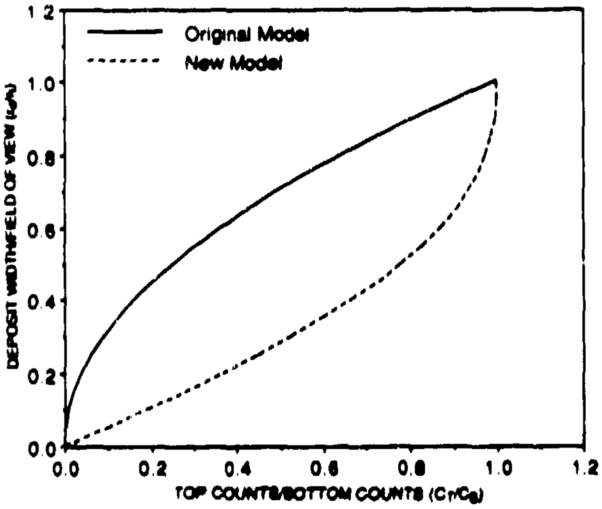

Fig. 3. Plot of the count rate ratio vs. the width ratio (deposit width to diameter of detector field of view) for the Los Alamos width model (new) and the Rocky Flats width model (original). The original model assumes a flat detector response and a circular deposit concentric with the detector field of view. The new model assumes a triangular detector response and a constant-width deposit across the detector field of view.

Table I. HEU Foil Specifications

Identification Number	Thickness (g/cm^2 ± 1 σ)	Effective* ^{235}U Thickness (g/cm^2)
1	0.03588 ± .0032	0.03256
2	0.02743 ± .0020	0.02504
3	0.02726 ± .0018	0.02490
4	0.03534 ± .0026	0.03209
5	0.04844 ± .0019	0.04356
6	0.05047 ± .0020	0.04532
7	0.05114 ± .0018	0.04590
8	0.04781 ± .0024	0.04302
9	0.05093 ± .0016	0.04572
10	0.05006 ± .0017	0.04497

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*Corrected for self-attenuation of the 186-keV gamma ray.

Measurement System Calibration

The gamma-ray spectrometry system was calibrated in the manner prescribed for generalized geometry holdup measurements.[1] Measurements were made using a sodium iodide detector 2.54 cm in diameter and 1.27 cm thick. A cylindrical lead shield around the detector reduced background radiation, and a tungsten collimator 1.9 cm in diameter and 3.8 cm deep restricted the detector's field of view to the same solid angle viewed by the bismuth germanate detectors used at Rocky Flats. A portable multichannel analyzer was set up to acquire a 0 to ~300 keV spectrum in 512 channels of memory. Regions of interest were set around the 60 keV peak from a ^{241}Am reference source, the 186-keV peak from the decay of ^{235}U, and a background region between 227 and 278 keV. Before measuring the detector's radial response to a ^{235}U standard, we made a 1000-s background measurement. We made 300-s radial response measurements at 2-cm intervals along a perpendicular to the detector axis, 40 cm from the detector. The normalized radial response curve is shown in Fig. 2. The calculated effective length and area were 17.37 cm and 308 cm^2, respectively. The area-source calibration constant was 0.0007248 g-s/cm^2.

Simulated Holdup Deposit Measurements

Pipes 14, 20, 24, and 27 cm in diameter were selected to simulate exhaust ducts. Foils were laid in the bottoms of the pipes parallel to their axes. By laying the foils adjacent to each other, holdup deposits up to 30 cm wide (depending on pipe diameter) were simulated. The detector was positioned 5.08 cm from the bottom of the pipe looking vertically upward for a bottom measurement. The background-subtracted count rate in the 186-keV region of interest was recorded. Then a spectrum was acquired with the detector positioned diametrically opposite to the first position, also 5.08 cm from the pipe. The paired measurements were repeated for as many different foil combinations as could be accommodated in each pipe.

In addition, data were taken with foils held flat by a Lexan cover plate. Measurements from 5.08 cm above the foils were made to simulate bottom measurements, and spectra were acquired from 21 to 34 cm above the foils to simulate top measurements. Care was taken to assure that the foil(s) filled the detector field of view during the simulated bottom measurements and that they did not fill the field of view during the "top" measurements.

Results and Discussion

The top-to-bottom count rate ratios were computed for each measurement pair. The ratio of the corresponding actual deposit width and geometric field of view is plotted as a function of count rate ratio in Fig. 4. The width ratio predicted in equation (3) typically errs from the actual ratio of deposit width to field-of-view radius by less than 10%. Note the relative insensitivity to deposit curvature: the model reliably predicts deposit width regardless of pipe diameter and even if the deposit is flat.

Having demonstrated that the new width model adequately predicts the width of relatively uniform deposits, it is useful to determine the accuracy with which holdup mass per unit length can be estimated. This is done by computing the mass per unit area using the bottom count rate and the area calibration constant, and multiplying by the deposit width determined by solving Eq. (3) iteratively at the corresponding count rate ratio. Results are shown in Fig. 5. For purposes of comparison in Fig. 5, the same date are interpreted using the original Rocky Flats approach, which predicts deposit width from Eq. (1) and computes an area's density using the top count rate. Note that the large overestimate of deposit width evident in Fig. 3 is partially offset by the underestimate of areal density that results from using C_T to calculate it, even when the detector's field of view is not filled.

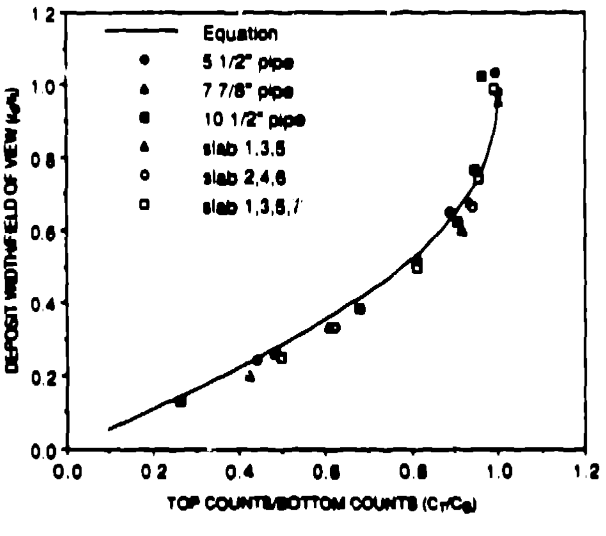

Fig. 4. Plot of the count rate ratio vs. the width ratio for the Los Alamos width model. Data were taken with 93%-enriched uranium foils arrayed in pipes of three different diameters and also laid on a flat surface. The distance of the detector to "deposit" in "ducts" was the pipe's outside diameter plus 2 in. less one pipe-wall thickness. Distance of detector to foils laid flat was 21 cm for "slab 1,3,5" data; 25 cm for "slab 2,4,6"; and 34 cm for "slab 1,3,5,7."

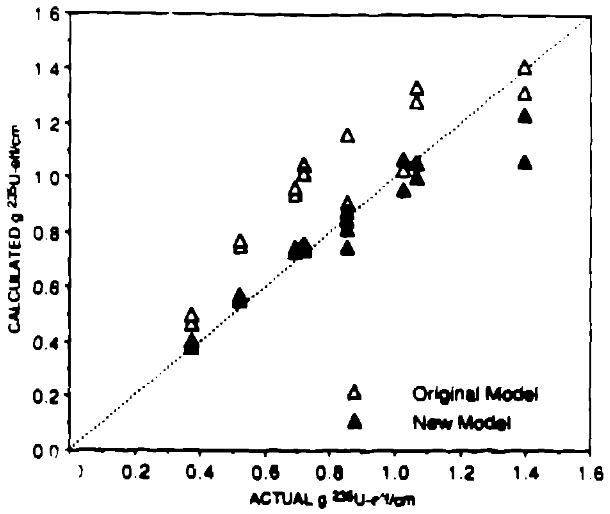

Fig. 5. Comparison of calculated vs. actual effective-mass per unit length of ^{235}U. Width results from the Rocky Flats model were multiplied by the mass per unit area derived from the top count rate to give the "original model" results. "New model" results were obtained using the Los Alamos width model and the mass per unit area derived from the bottom count rate. The presence of multiple data points at each actual ^{235}U mass per unit length is attributable to different pipe diameters.

Because the foils are not all the same thickness, it is difficult to configure a simulated deposit that has a uniform thickness across it. The top detector is relatively insensitive to this problem because it sees a larger area and tends to register an "average" count rate from the entire foil array. If an unusually thin or thick foil is in the center of the array, where the "bottom detector" views it, then a biased estimate of either the deposit width or the areal density can result. The anomalous data points in Fig. 5 are probably attributable to this phenomenon.

Further tests of the effects of nonuniform deposit thickness were performed by stacking foils on top of each other. When the deposit was thinner down the center than along its edges, the top-to-bottom count rate ratio was observed to be greater than one. Neither width model is applicable in this case. Applying either model with the assumption that $C_T/C_B = 1$ results in a large negative bias in the holdup estimate. Inverting the geometry to postulate that the deposit is in the top of the duct makes the count ratio a fraction of unity, but gives even poorer results.

When the foil array was thicker in the center than it was on the edges, C_T/C_B was observed to be a fraction of unity. Application of the Los Alamos model resulted in a 20% low bias in the holdup estimate. The Rocky Flats method gave holdup estimates that were 10% to 15% high.

CONCLUSIONS

For holdup deposits that reside as uniform strips in the bottom of the duct, the new width model described above provides a reliable method for interpreting data taken at contact using the Rocky Flats strategy of positioning the detector at the top and bottom of the duct. The sensitivity of this technique to nonuniform deposit thicknesses, however, limits its application to those cases in which reasonable assurance of deposit uniformity exists. Where $C_T/C_B \geq 1$, or where there is evidence of a nonuniform deposit geometry, it is preferable to withdraw the detector from contact with the duct to assure a line-source geometry.

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