

STANDARDS FOR HOLDUP MEASUREMENT *

M.S. Zucker
Brookhaven National Laboratory
Upton, New York 11973

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Abstract

Holdup measurements,¹ needed for material balance, depend intensively on standards and on interpretation of the calibration procedure. More than other measurements, the calibration procedure using the standard becomes part of the standard. Standards practical for field use and calibration techniques have been developed. While accuracy in holdup measurements is comparatively poor, avoidance of bias is a necessary goal.

General Remarks

The reduction of counting data obtained from holdup measurements to equivalent amounts of special nuclear material would be nearly impossible without standards. Their main application is calibration of instruments used in measurement geometries for which calculations from first principles (allowing for attenuation, etc.) would be too uncertain or laborious.

Holdup measurements individually are often inaccurate for various reasons, such as differences in and non-uniformity of the deposited material, unfavorable geometries, etc., leading to a large uncertainty in the total amount. Since the aggregate amounts may be significant, it is important nevertheless to minimize systematic errors. This in turn can be accomplished by calibrating the counting equipment under the most realistic conditions possible. Then, even if individual measurements are incorrect due to some vagary such as the deposition of material being non-uniform, the measurements will be representative "on the average", and the total will not differ systematically from the true mean.

A key element in preparing for holdup measurements has therefore been the development of standards and particular way of using them to effect hopefully realistic simulations of typical measurement situations and geometries. The latter includes a few simple physical and mathematical techniques which allow the use of standards to simulate situations beyond what might be expected to be limitations, because of size.

Although it might not seem so at first glance, most of the objects found in nuclear manufacturing plants, particularly if process equipment is removed, can be idealized geometrically as regards the nuclear radiation coming from deposits of special nuclear material and treated quite realistically as flat surfaces (a wall, ceiling, etc.), or hollow cylinders of round or rectangular cross sections (duct work and piping, etc.). For these situations, the standards which are appropriate are those which simulate area sources of radiation. Appreciable effort has gone into their

design and fabrication. These sources, prepared from representative types of special nuclear material, are used together with samples of duct work or piping either donated by the plant or procured commercially, to calibrate the instruments used in the measurements.

In cases where it is not practical to make up area standards of the required dimensions, they can be simulated using a smaller area source together with a particular measurement technique and mathematical treatment.

In some of this calibration work it is necessary to assume that an inverse square law (for the fall-off of the intensity of radiation as a function of distance) be applicable. The conditions for this have to be examined.

Some situations in holdup measurement require that calibration measurements together with a standard mathematical procedure be considered as the calibration standard.

After all of the obvious places (glove boxes, furnaces, etc.) for holdup have been explored and the material recovered, the bulk of the remaining holdup seems to be in the form of relatively low concentrations of material, however spread over rather large areas (floors, walls, and ceilings) so that the aggregate amount may be impressively large. From the safeguards point of view, these area concentrations are considered rather important, because while process equipment can be cleaned out by conventional techniques, and even removed for scrap or burial, the floors, etc., of the plant remain behind. There is then not only a question of how much material is present, but also how easily it can be "mined" surreptitiously at a future date, and so to what extent it must be guarded. Probably for all those reasons the most effort has gone into dealing with areas. This has resulted in the development of standard area sources, instrumentation intended for assaying areas, and a few calculational methods for areas.

Manufacture and Characterization of Area Standards

Area source standards, that is to say, a standard made to simulate nuclear material thinly deposited over an area as far as the radiation emitted goes, have been made two different ways as outlined below. The aim in either case to produce a thin flat object on which a known amount of oxide powder has been uniformly distributed over a known area with the nuclear material safely contained.

The first method is based on the availability of thin transparent sheet plastic which has one side coated with adhesive, intended for

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laminating to documents, etc. The adhesive is protected before use with a sheet of material which can readily be peeled off.

Using a straight edge and razor blade a rectangular area of the backing material is cut out. This leaves a rectangular area of adhesive surface exposed lying within a border where the backing material still remains. This is weighed with mg precision and accuracy. The powdered oxide is put onto the adhesive using an artist's brush. This is done while the plastic is illuminated from behind, so the uniformity of deposition can be judged. Excess material is shaken off, and the plastic with oxide, and the remaining protective material, carefully weighed again to find the deposited material. The border of backing material is then peeled off, and another sheet of the adhesive plastic applied over the first so that the adhesive surfaces match. In this way the area with the oxide material is sealed within a border free of active nuclear material. After checking for outer surface contamination, the laminated object is bagged out of the glove box in polyethylene, heat sealed to just fit.

It has been found possible to make deposition of oxide of 0.01 to 0.1 g/cm², over areas of $\sqrt{200}$ to $\sqrt{500}$ cm², which give sufficient count rates for holdup measurement purposes. The amount of oxide that can be deposited depends on the "stickiness" of the adhesive and the grain size of the oxide. The latter is often too fine, in which case the sticking power of the backing will be used up without achieving a sufficient loading. In such cases, a small amount of cellulose dissolved in acetone (or some similar glue) can be mixed with the powder, which is then dried, crumbled, and sieved to obtain a coarser grain size. It has proven more difficult to make the deposit uniform. This is, as before, checked visually by illuminating the sheet from behind. Densities in the desired range will show up as translucent gray, from light to fairly opaque. The worst of the group are discarded. The self absorption factor, measured by the standard technique for radioactive material (see below) is of the order of 1.02 or 1.03.

A second method was developed to overcome problems anticipated with the first, that they might not prove rugged enough, the resulting sources could only be bent in one direction, and that the loading was limited by the adhesive and grain size. In this method, a weighed amount of oxide is mixed with silicone rubber monomer, catalyzed, and poured over a previously prepared molded sheet of silicone rubber without nuclear material. The layer with oxide can be poured to within say, 1 cm of the edge of the oxide free sheet. After this hardens, another layer of silicone rubber is poured over the previous two sheets, thereby sealing both sides of the oxide containing sheet with sheets of oxide free silicone rubber, and leaving an oxide free border surrounding the active material. The result is a rubber mat, about 5 mm thick, which meets the above objections to the first type of area standard. It

is however somewhat clumsier to use and has a larger self absorption factor, $\sqrt{1.15}$ or 1.20. Since the mat is not transparent, the uniformity is checked with a collimated NaI detector. The only trick involved in the manufacture is to get out gas bubbles from the viscous liquid (by exposing the mixture to reduced atmospheric pressure) and to flatten the unpolymerized liquid (done by letting the rubber cure while held in a shallow metal tray coated with parting compound, and slamming the tray down flat on a table a few times).

It is thought that the amount of material per unit area in either of these area standards is accurate to about 3%. The uniformity is probably no better usually than $\pm 15\%$. However, as typically used, the detector will view a large enough area of such a source so that local fluctuations in density will average out.

Samples of duct work, piping, tubing, etc., of the type used in a given plant are also useful in conjunction with the standards described above to determine the attenuation of radiation coming from nuclear material deposited on the inside under the actual measurement conditions. The area standards are inserted inside the sample pipe or duct, etc., so as to conform to the interior surface and cover it within the solid angle viewed by the detector (both near and far walls). There has been only one test of how well this procedure works. In this case the manufacturer expressed doubt at how much material was indicated by measurements based on such a calibration procedure. The duct work was dismantled, and the material taken out and weighed, and, perhaps fortuitously, there was agreement within $\sqrt{10\%}$. No tests under laboratory conditions have been made.

The area sources can also be held against the inside surface of glove box walls to measure the actual absorption of the walls. Radiation from the outer walls or objects within the glove box can be shielded from the detector with sheet lead placed in back of the area source.

Another useful source is one made of a disc of uranium metal foil of known weight ($\sqrt{2.5}$ gU), area ($\sqrt{20}$ cm²), isotopic composition (usually $\sqrt{93\%}$ enriched), and self absorption factor ($\sqrt{1.06}$), laminated between plastic sheet for protection. The foil used to make such sources was originally intended for use in critical facility studies, and the small amount (or unirradiated foil) needed might be obtained from institutions that have done such work, or from the U.S. DOE laboratory ORNL. A disc can be stamped out of the foil using a "cookie cutter" made of a steel tube, one end of which has the edge sharpened.

Such a disc can be used as an element of area with a particular density of nuclear material. Placed far enough away from a gamma detector it can simulate a point source of characteristic (186 keV) U²³⁵ radiation. With the weight and isotopes known, the disc can be

used to calibrate the detector if the self absorption factor (SAF) is measured,² illustrated in Figure 1. A lead mask (1 cm thick) is used to define an area on the "unknown" source for which the SAF is to be measured and also, to rigidly control distances from the detector. The unknown is counted alone, (A), then an auxiliary source of U²³⁵ radiation is counted alone, (B), then both together, (C). Then, writing $\exp(-x) = (C)-(A)/(B)$, the SAF would be $x/(1-\exp(-x))$, where $x=μt$ is the product of linear absorption coefficient and thickness for the unknown. Multiplying the count rate of a detector viewing this source by the SAF would give the count rate if there were no self absorption, thus connecting the count rate with the amount of U²³⁵ in the source.

The SAF can be measured to at least three significant figures. There is no point in doing any better, since it is based on the assumption of an idealized plane parallel geometry. The amount of material in the disc is easily known to 0.01g; the isotopics to almost 3 significant figures.

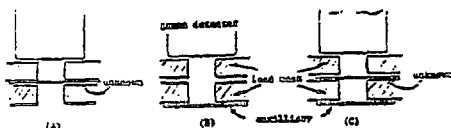


FIG. 1 Illustrating the absorption factor determination

Walls and ceilings have been assayed with gamma detectors calibrated using area sources of the type described above, or by simulating an area.

In using an area source for calibration, the inverse square law is invoked. The detector is held perpendicular to the area source at such a distance that the collimation allows exposure just to the active area of the standard, and yet is far enough away so that the detector approximates an ideal point detector. There is an obvious conflict in the two requirements, since it is hard to make uniform area sources large enough to make a typical NaI detector look like a point. (The development of high resolution NaI detectors consisting of 18 mm DxdH crystals mounted on 18mm photomultiplier tubes to take the place of the previous standard 50 mmD detector has made this easier.)³ A test is to measure the count rate at two different distances. For the ideal case of a point detector and an infinite plane source, the count rates should be the same. In practice, 10% agreement is taken as adequate.

Another way of simulating an area source which avoids some of the above problems, but introduces some new ones perhaps harder to evaluate is the following: The detector axis is held perpendicular to a clean (radioactively speaking) wall, about 1 m away. A disc source of the type described previously is then moved on the wall along a line passing through the point of intersection of the detector axis and the wall.

Counting the disc in the successive position generates the response of the detector to material deposited on the plane as a function of radius from the on axis position. This can in turn be related to how the detector would respond to an infinite plane source having the same amount of nuclear material per unit area as the disc.

The particular algorithm used depends on the details of the experimental set up. For an example, suppose the disc is moved in increments equal to its own diameter. Then if C_i is used to denote the count obtained with the disc in position i , where $i=0$ corresponds to being on the detector axis, and $i=1,2,3...$ refers to the center of the disc being respectively 1 diameter, 2 diameters, etc. removed from the detector axis, then the summation

$$S = C_0 + 8 \sum_{i=1}^{\infty} i C_i$$

is the count rate if the whole wall were covered with a source having a density of nuclear material the same as the disc. Allowing for the self absorption factor of the disc, the count rate would be $S' = (SAF)S$.

The statistical precision of this type of calibration is quite good (1%). However, there have not been any experiments to determine how accurately measurements made in the field represent the actual holdup. On one occasion however, this procedure was tested by using the detector calibrated this way to assay an area source standard of the type whose manufacture was described above. The agreement was 10%.

The above procedure seems reasonable when applied to walls and ceilings, where the deposits are on the surface. (An exception is occasionally found with walls near process equipment where liquids have leaked.) In the case of plant floors nuclear material generally seems to have diffused into the concrete carried there by successive spills, and perhaps even aided by clean up attempts. In this case the floor areas are divided up into a grid of 50 or 100 cm squares, and each one scanned with a collimated gamma detector (calibrated with an area standard) moved parallel to the floor. These scans are used as indications of the concentrations of nuclear material in the concrete. The area scans are used to assign preliminary values, which in turn are used as a guide for drilling bore holes in the concrete according to a reasonable sampling pattern. The holes are drilled to a depth such that mea-

surement indicates no further activity is detected (usually $\sqrt{7.5}$ cm). The dust is collected with about 99% efficiency using a specially developed suction apparatus which separates the air stream from the entrained dust using a cyclone separator principle.¹ The air is passed out through absolute filters, while the dust is caused to collect in vials suitable for counting in a NaI crystal well counter. After a gamma assay for each vial, they are sent to the U.S. DOE Analytical Laboratory NB¹ for element and isotope analysis.

An attempt is then made to correlate the amount of material recovered from each bore hole with the surface scans for that area. This correlation is then used to establish the probable total for the whole area. An obvious reason for such a correlation to fail is the different amounts of shielding due to different degrees of penetration of the concrete by uranium bearing solutions in different areas. However, the fact that there is a large amount of sampling done hopefully causes the correlation to be representative on the average. There have been no studies to indicate the accuracy of the procedure, except that part having to do with collecting the dust: A sample of concrete drill dust, collected, weighed, spread out again, collected again, etc., indicated of the order of a 1% loss on the average.

Conclusions

It may be inferred from the above that there is a difference between holdup standards and ordinary standards. The standards generally used to calibrate nuclear material assay equipment are very similar or even nominally identical to the items being assayed. In contrast, the standards used for holdup measurement can be said at best to only simulate the actual physical measurement after some plausible but highly idealized mathematical model is applied to the analysis of the calibration data.

In a sense then, the calibration standard includes not only some radiation source, but also a measurement procedure and a way of analyzing the data from it. It would seem all the more important, this being the case, to test these methods. As indicated above, this has not been done for the most part. Partly this is probably because the funding authorities in the U.S. have tended to weigh the cost of such research against the value of the material involved (admittedly small compared to the value of special nuclear material to which other NDA techniques can be applied). What should perhaps be considered however, is the safeguards value of holdup measurements, since that is the final item in the material balance for a campaign or plant operation.

Another feature of holdup measurements which should be looked into is the best strategy for doing such an assay, factoring in not only the likely amounts of material (which is done now to some extent on the basis of a pre-

liminary survey), but also such quantities as strategic value of the material, and ease of recovery, etc.

Reference

1. M.S. Zucker, et al., Holdup Measurement for Nuclear Fuel Manufacturing Plants, 1981, INMM Proceedings.
2. We are indebted to P. Colmann of the BNL Reactor Division for a discussion on the practical aspects of making this measurement.
3. M.S. Zucker, et al., "A New Survey Assay Meter for Portable Applications" Proceedings 1st Annual ESARDA Symposium, April 25-27, 1979.