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Holdup Measurement For Nuclear-Fuel Manufacturing Plants

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Abstract

The assay of nuclear material holdup in fuel manufacturing plants is a laborious but often necessary part of completing the material balance. A range of instruments, standards, and a methodology for assaying holdup has been developed. The objectives of holdup measurement are ascertaining the amount, distribution, and how firmly fixed the SNM is. The purposes are reconciliation of material unbalance during or after a manufacturing campaign or plant decommissioning, to decide security requirements, or whether further recovery efforts are justified.

Introduction

The term "holdup" as applied in the nuclear industry refers to the various chemical forms of uranium or plutonium or perhaps other "special nuclear material" (SNM) which become deposited in process equipment, duct work, walls, floors and ceilings of a nuclear manufacturing plant during its operation. Usually, this material is in a form which is not easily removeable, i.e., "fixed," otherwise it would have been recovered as a part of normal, periodic, cleanup operations.

In some cases, however, surprising quantities of removeable material have been discovered. The typical place for this to happen is part of the process equipment considered inaccessible under the impetus of routine clean up operations, or where it was not realized that individual repetitive actions, each contributing negligible amounts of material over a long enough operating history, would result in an appreciable accumulation.

The objectives of holdup measurement are ascertaining the amount, distribution, and how firmly fixed the SNM is. The purposes are to help reconcile material unbalance during or after a manufacturing campaign or plant decommissioning, to decide how much security is needed from the safeguards standpoint, and whether further recovery efforts are justified from either safeguards, economics, or health and safety considerations.

The degree that nuclear material is considered "fixed" may be a fairly objective matter as regards health and safety considerations, but is perhaps more a matter of judgement with respect to safeguards. A working definition is that SNM is fixed when the amount present, as indicated by radioactivity measurements, approaches an asymptotic value upon repeated application of appropriate cleaning procedures. The word "appropriate" is what needs interpretation on occasion. It can range from a scrubbing with detergent to tearing up concrete.

A feature of holdup measurement is that the sum total of the material uncovered in the search is a monotonically increasing function of the number of measurements made. There is therefore a built-in systematic error, a bias in that the amount of material which can be reported as found must be less than what is actually there, by some unknown amount. This leads to a basic conflict and frustration in holdup measurements about allocating resources to maximize the amount of nuclear material uncovered without getting too far past the point of diminishing returns and prohibitive expense.

Another aspect of holdup measurements is the extent to which the fortune of the facility may be perceived by the management as tied to the outcome of holdup measurements. The management may hope for the holdup to be high, because that would help explain material previously unaccounted for. Or, it may be in management's interest that the figure be low and or fixed so that a smaller guard force be necessary for a decommissioned and non productive facility, or so that the facility can be converted to other use. There can therefore be substantial financial interest in the outcome of a holdup measurement.

There are certain qualitative differences to holdup measurements compared to most other nuclear based measurements made under laboratory or even factory or field circumstances. These factors generally significantly reduce the precision and accuracy that can be expected: The mea-

surement situations tend to be poorly controlled or characterized as to geometry and the nuclear material and matrices involved. The nuclear material tends to be diffuse, and it may be difficult or impossible to isolate objects being assayed from radiation from other objects.

As a general rule, ±5% or ±10% measurements are exceptional, the more usual being 25% or even ±50%. Usually one is resigned to such results and hopes at least the mean value attained will not reflect a bias due to poor calibration, some instrumental effect, or an artifact of the approximations used in data reduction, etc.

There is, much more so than usual in physical measurements, room for honest differences of opinion in interpreting the raw data and reducing it to an equivalent amount of nuclear material.

Statistical considerations, for example in deciding sample size, play a secondary role in determining holdup measurement strategies, compared to other NDA measurements. Rather, the emphasis seems to be on maximizing the amount of material uncovered subject to the time and manpower available. Probably the basic decision is which areas or pieces of process equipment are likely to yield the most significant amounts of material. Sampling tends to approach 100% because of the fear that some "hot spot" might be overlooked. The practical goal is in fact to do everything at least once lightly and then examine promising places in more detail.

The burden of holdup measurements on personnel and equipment.

Holdup measurements often have to be made under conditions unfavorable for personnel and equipment. Manufacturing plants in or approaching a shutdown condition frequently have their air conditioning systems off and therefore too warm or cold for comfort. Some plants in operation can be as bad or worse, with excessive heat from furnaces, and noise from pumps and ventillation ductwork. The noise level can be so bad that hearing protection ear muffs have been used to reduce operator fatigue, though these can be uncomfortable in areas of high heat and humidity.

The risk of personnel and equipment becoming contaminated with radioactive material is enhanced during holdup measurements. Areas that were not available for routine housekeeping operation during plant operation, allowing accumulations of nuclear materials dust, have to be visited. Certain of the operations, such as drilling for concrete samples, raise dust and spread contamination. Extra protective clothing, including masks have to be worn sometimes because of these hazards. In addition to radiological hazards, there often are physical hazards attendant on climbing in and around elevated structures, to view the tops of glove boxes, ductwork, and piping. In addition holdup assay often involves seemingly endless repetitious measurements. The discomfort to personnel making holdup measurements can be severe enough so that even highly motivated individuals might tend to compromise on the quality and thoroughness of measurements.

This generally unfavorable and unpleasant environment that holdup measurements are made in makes it even more important than is usually the case for the measurement equipment to be simple to use, easy to interpret, stable against temperature and count rate effects, self-contained, rugged and portable. It should also be as sealed as practical against infiltration by radioactive dusts and with surface finishes that can withstand being washed down with strong detergents. Since an inordinate amount of time and effort often must be spent in decontaminating equipment, sheet polyethylene is often used to wrap the equipment for protection. Electronic equipment which is push-button operated is more suitable for this treatment than that requiring rotary or slide switches. Fortunately this is the trend in modern equipment especially that which is microprocessor based. With polyethylene or similar wrapping, the build up of excessive heat in the electronics has to be watched. Here again, the modern trend to lower power solid state devices helps. Polyethylene sleeving is commercially available and can be used to protect cables and power cords; in some cases though it has proved expeditious to consider the cables as expendible supplies. Electric drill motors often get contaminated internally due to the stream of air drawn through them for cooling when they are operating. Unfortunately except perhaps for very expensive types, they are usually not worth the effort it takes to decontaminate them internally, and have to be abandoned or donated to the facility for use there. Wheeled equipment can be protected in large measure by wrapping tape around the wheels as is standard practice anyway on entering a nuclear facility. Devices with reentrant cavities for counting packages or samples can easily be lined with polyethylene sheet or bags.

The setting for holdup measurements

Nuclear fuel manufacturing lines typically consist of a series of glove boxes, interconnected with communicating tunnels as far as possible to avoid bag-in or -out operations. The whole system is kept at a negative pressure with respect to the room. The tunnels may be short enough to allow a manual pass through, or employ some sort of mechanical conveyor. Furnaces may be part of the glove box sequence, or may stand alone.

The chief difference between a production line handling plutonium and one for uranium seems to be that in the case of plutonium much more effort is made to contain every part of the process in one continuous system. For example, a uranium process line might very well have parts of the process which are inherently hard to confine to even a large glove box, stand

alone. In particular there is liquid phase chemical processing equipment of various types and purposes which often take the form of elongated vertical columns. In a plutonium process line, even these awkward shapes may be housed in very large multistory glove boxes. In either case, there will be piping conveying chemical solutions of nuclear material connecting or internal to the glove boxes.

The slightly negative internal pressure is maintained through the use of ductwork connected through a series of filters to a building exhaust. More than one filter is used in order to protect against a failure.

The above brief description sets the scene for the development of holdup in a process line. The chemical process equipment will occasionally spring leaks at pipe joints, value stems, transfer pumps, gasket surfaces, etc. This tendancy is aided by the corrosive nature of the nuclear material solutions and the fact that the lines are often flushed out with even more corrosive chemicals to remove residues of the original solutions.

Moreover, to begin with, the process lines have often been designed and built to be amortised in a relatively short time, consistent with practice in the non-nuclear segment of the chemical industry, and perhaps as is appropriate in an industry which has labored under technical and economic uncertainties from its inception.

Special nuclear material in the glove box line often is in the form of oxide, either as a powder or compressed sintered powder. Even in the sintered form, abrasion of the pellets against one another or their conveyances causes powder to be generated. There is an appreciable flow of air within the glove box due to the negative pressure system, allowing the dust to become entrained thru elutriation. A certain portion of this ends up as a light coating over all surfaces of the box, the rest is passed into the duct work, where it is deposited along the duct or trapped in the absolute filters.

Some times through carelessness in adjustment of airflow, higher velocities than needed for negative pressure are allowed to exist in parts of glove box systems resulting in excessive entrainment of powder in the air stream.

The powder is deposited in duct work in a not necessarily uniform manner, and for the most part much as one would expect from elementary considerations, e.g., a horizontal length of duct will tend to have most material deposited in the lower half. Other places where material tends to be concentrated are near where the duct work undergoes a turn with a small radius, or there is a sharp change in cross section. One can speculate that devices operating on principles similar to cyclone separators might be inserted in a duct work system after particular glove box stations that tend to produce more than usual amounts of dust, in order to recover material carried by the air stream. These separators would have to be monitored for criticality. This might be preferable to periodic cleanouts of duct work and converting the absolute filters to ash.

Automatic conveyor systems, e.g. for carrying trays of pellets through tunnels or furnaces, are another source of holdup, as pellets or fragments are occasionally jostled out of the trays. These places are less accessible than most in the glove box system, hence unlikely to be thoroughly inspected on a periodic basis.

Calcining ovens are another place where there can be surprising accumulations of nuclear material. A problem with them, as with sintering furnaces, is that the walls are thick and shield the material against external detection.

Special nuclear material has a tendency to plate out of solution on the walls of pipes, chemical columns, and on items such as Raschig rings, giving rise to yet another form of holdup.

The actual layout of the individual pieces of equipment and details of the process can furnish clues as to where concentrations of holdup might be found. Employees are often knowledgeable about the history of the facility, such as where certain process equipment had been located, or more to the point, where particular spills occurred.

General problems encountered in holdup measurements

The interpretation of data taken with gamma or neutron sensitive instruments in terms of the amount of nuclear material that gave rise to the radiation, is heavily dependent on idealized models and standards. It is often convenient to consider individual items as behaving essentially as either point, or line, or cylindrical, or plane (area) sources of radioactive emissions.

Holdup in the form of material deposited over areas often make the biggest contribution to holdup; the material per unit area may be miniscule, but prorated over large areas, such as glove box sides, or plant walls, may amount to sizeable quantities of material. A useful concept applicable in the case of areas is the fact that because of the inverse square law that nuclear radiation obeys (neglecting absorption), a point detector will count at the same rate no matter how far it is from a uniform plane source. This is because as the distance of the detector from the plane changes, the change in intensity due to the inverse square law dependence of the radiation is just compensated by the change in that portion of the source area that the detector views, keeping the total count rate the same.

This is often a good approximation in practice. The degree to which it is in the case of a particular (finite sized) detector housed in a particular collimator can be evaluated by comparing a series of counts taken as the detector is moved to various distances from an artificial plane source of radiation. The source should be uniform, and of an area large enough that the area viewed by the collimator lies within the boundaries of the source. The count rate should, under these circumstances, remain sensibly constant (within, say, 10%) as the detector to plane source distance is varied. Knowing the amount of nuclear material per unit area in the plane source (and the self absorption factor if it is appreciable) will serve to calibrate the detector for viewing large areas of wall or ceiling if the deposition of material is uniform. In order to implement this in a situation where the surface deposition may change from one area to another the solid angle of the collimator detector system must also be known so that the viewed area be kept reasonably uniform. Preliminary surveys of the wall can be made to test the uniformity. The solid angle can be taken with sufficient accuracy to be the area of the aperture in front of the collimator divided by the square of the distance from the effective center of the detector's to the center of the aperture. The detectors effective center can be determined empirically for a given energy of radiation by finding (by trial and error or otherwise) that length which must be added to the distance between a geometrically small radiation source of the proper energy and the front face of the detector in order for the count rate to appear to obey the universe square law. This means, e.g., that multiplying the count rate by the square of the adjusted distance between source and detector gives a constant, independent of the distance. The effective center is on the axis of the detector somewhere between the front face and midpoint of the axial length of the crystal.

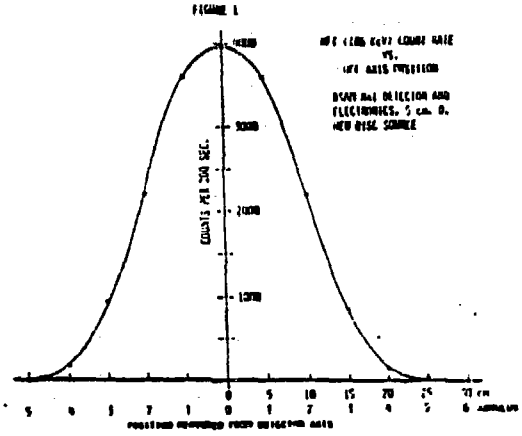
In cases where the deposit is not uniform, the assay will result in a representative albeit crude value, but one which is not biased, i.e., there will be compensation on the average for all the areas measured.

In case adequate area sources are not available, it is still possible to synethesize the equivalent from a geometrically small source. The procedure involves finding the response of the collimated detector to a source of radiation as a function of its distance (radius) off the axis of the detector in a plane perpendicular to the axis, and using this response to infer what the count rate of the detector would be if it faced an infinite plane source with the same density of radiation per unit area as the small source.

There are several ways of implementing this. The following is one prescription or algorithm which has been used. The only equipment needed is the collimated detector and electronics to be used in the area measurements, a relatively small (geometrically) source having the proper isotope (e.g., a 5 cm D x ¼g, HEU foil disk source in the case of uranium holdup measurements), and a meter stick.

The collimated detector is lined up perpendicular to a wall which is free of radioactivity. A scale is marked off on a radius from the point of intersection of the detector axis and the wall. A convenient interval for the scale is the diameter of the disc source. The disc source is then successively centered at each of the radial intervals and then assayed. As the disc is moved to further off axis positions, the count rate drops monotonically. Figure 1 shows such data.

FIGURE 1



The radial positions may each be considered as denoting an annulus of area on the wall about the point of intersection of the collimator axis and the wall. The annulus immediately about this point (really a circle) has an area πr_o^2, where r_o is the radius of the disk; call this one the zeroth annulus. The next annulus (the 1st) extends between r_o and $3r_o$ and has an area $\pi(3^2r_o^2-r_o^2) = 8\pi r_o^2$, the next (2nd annulus) has an area $\pi(5^2r_o^2-3^2r_o^2) = 16\pi r_o^2$, *etc.*; the general formula for the area of the ith annulus, with $i > 1$ is $8\pi r_o^2 i$. With this particular choice of radial interval equal to twice the disc radius, the ratio of the areas of the successive annuli to the disc area is as $1,8,16,24,32,\ldots,8i,\ldots$. If the whole wall were covered with a uniform source having the same density as the disc source, the collimated detector would therefore have a count rate given by the sum of the count rates just observed each weighted by the ratio of the area of the respective annulus to the area of the disc. This count rate should be corrected by the self-absorption factor for the disc. The following table indicates the mechanics of this process with actual data.

Table I. Calibration data for a particular collimated NaI detector with associated electronics

Annulus	C_i counts in 200s at annulus i	Annulus to source area ratio R_i	Product R_iC_i	Product $R_i^2C_i$
0	3930	1	3930	3930
1	3604	8	28832	230656
2	2234	16	35744	571904
3	896	24	21504	516096
4	178	32	5696	182272
5	18	40	720	28800
6	0	48	0	0
			96446	1533678
			$= \sum_i R_iC_i$	$= \sum_i R_i^2C_i$

$\sigma(\sum_i R_iC_i) = (\sum_i R_i^2C_i)^{1/2} = 1238.4$

$\frac{\sigma(\sum_i R_iC_i)}{\sum_i R_iC_i} = 0.0128 \approx 1.3\%$ relative (statistical) uncertainty

The distance of the detector from the wall is not crucial, 1-2 m is enough for the detector to be a good approproximation to a point detector. The column giving the product $R_i^2C_i$ calculates the variance of each of the entries, leading to a statistical uncertainty in the calibration of ~1.3%. Since there are systematic errors involved in this which are probably at least of that order (for example the finite width of the annuli, with the implicit assumption that the contribution from each part of the annulus is constant with radius), there is no point in striving for greater accuracy. In this particular example, the disc contained 3.640 g U^{235}, a self absorption factor of 1.365, and an area of 20.27 cm^2. Thus, the calibration factor for this particular instrument is

$$\frac{96{,}446 \text{ counts}}{200 \text{ sec}} \times 1.365 \times \frac{1}{\frac{3.640 \text{ g } U^{235}}{20.27 \text{ cm}^2}}$$

$$= 3666 \text{ (cts/sec)}/(\text{g}U^{235}/\text{cm}^2)$$

This means e.g., that a uniform deposition of about 1 mg of U^{235} per cm^2 would produce a count rate in this detector of ~3.7 cts/s.

A problem that has risen occasionally in measurements of uranium holdup using the familiar two channel analyzer type of instrumentation (BSAM or ESAM) is the presence of radiation due to another nuclide (for example Th and daughters) besidea U^{235} and U^{238}. This causes a problem in that the proportion or fraction f of the upper channel (Ch2) that must be subtracted from the lower channel (Ch1) in order for the difference to represent just the U^{235} contributions to Ch1, was determined using just uranium, ideally U^{238} (or depleted U) alone. (If there were only uranium present, then the count due to U^{235} in Ch1 is $(U_1 - fU_2)$.) The presence of a background due to a third nuclide alters the value of f that should be used. Moreover, the amount of this background, hence the appropriate value of f, in various areas of the plant will change.

The two channel analysis system can however accommodate this additional background in the following way: Let $U_{1,2}$ be the contribution from uranium to $Ch_{1,2}$, and suppose that the contributions of the other nuclide to $Ch_{1,2}$ be $T_{1,2}$. Let the ratio (Ch_1/Ch_2) from a pure U source be u, and that from a pure "T" source be t. Suppose that for a particular measurement of holdup the values recorded in $Ch_{1,2}$ are $M_{1,2}$ respectively. Then assuming linear superposition $M_1 = U_1 + T_1$, $M_2 = U_2 + T_2$ and $(U_1/U_2) = u$, $(T_1/T_2) = t$.

These are four equations in the four unknowns $U_{1,2}$, $T_{1,2}$. The quantities of interest $U_{1,2}$ can be solved for:

$$U_1 = \frac{u(M_1 - tM_2)}{(u-t)} \qquad U_2 = \frac{M_1 - tM_2}{(u-t)}$$

Then the net count due to U^{235}, call it N_{235}, is given by

$$N_{235} = (U_1 - fU_2)$$

using the value for f found with a depleted uranium sample. To illustrate this with an actual case:

In a particular situation, background radiation was found to be coming from the material that the cement block wall was made of. Using a sample of the wall known to be free of uranium holdup, the ratio $t = T_1/T_2 = 0.27660$ was found. Using uranium alone of isotopic composition similar to that of the holdup, the ratio $u = (Ch_1/Ch_2) = 34.562$ was obtained. A depleted uranium sample gave a value $f = (Ch_2/Ch_1) = 0.49378$. Thus, $u-t = 34.285$, $u/(u-t) = 1.0081$. In a particular measurement, Ch_1 had a value $M_1 = 4359$, Ch_2 read 1307. Thus $M_1 - tM_2 = 4007.5$, $U_1 = 4039.9$ and $U_2 = 116.9$, giving a U^{235} contribution $N_{235} = 3950$, to which the calibration factor derived from a standard would have to be applied to determine the amount of U^{235} per unit area.

This type of calculation can of course be set up as a program in the repertory of a calculator such as the one with the BSAM III, see below.

Detectors, electronic instrumentation, and other measurement aids

The most useful instrument by far in holdup measurements is a collimated NaI-photomultiplier (gamma) detector coupled to a portable dual single channel analyzer. The optimum size of the

crystal is debatable. A larger diameter crystal-photomultiplier combination has more sensitivity but will require a much heavier collimator for a given background attenuation, an important consideration in a portable application. A thinner crystal, say ~10 mm will favor the characteristic 186 keV radiation of U^{235} over the 765 and 1001 keV radiation of U^{238} and therefore have a more favorable peak height to Compton background ratio, but will have poorer resolution other things being equal, and be inferior if it is desired to use it for more energetic gammas, such as Pu (the 380 keV complex), compared to say, a ~25mm thick crystal. We have found an 18mm D x 18mm H (3/4" x 3/4") crystal mounted on an 18 mm photomultiplier tube to be an overall best compromise.

In any case it is necessary that the NaI-photomultiplier detector gain control against temperature and count rate changes. Such detectors are now commercially available in all common sizes.

A NaI well crystal, say 5 cmD x 5cmH, with a hole no bigger than necessary to accomodate a small plastic sample vial, have been found quite useful for assay of powder samples, and to identify and measure nuclides in scrapings, debris, etc. Germanium diode detectors with wells are also commercially available and may be useful.

Hand held "intrinsic" (high- or hyper-purity) germanium detectors have been developed,[1] partly with holdup measurement in mind. Non-spilling liquid N_2 dewars allow these detectors to be oriented in any direction. Their resolution is good enough so that isotopic analysis of the holdup could in principle be done in situ, though this has not yet at this writing been done. An intrinsic detector can be useful even for a two channel analysis in order to separate interfering peaks which cannot be resolved with NaI p.m. detectors, such as when recycled uranium is being assayed. Even for other work, the fact that the resolution is superior to NaI may give a better signal to background ratio that more than makes up for a lack of sensitivity compared to NaI-p.m. detectors.

Portable neutron detectors such as the "SNAP"[2] have been developed which can be hand carried, and may be useful to assaying plutonium in the form of holdup. They are only moderately directional (a front-to-back ratio of ~6:1), and it maybe difficult to interpret the count since neutron production from Pu depends not only on the isotopics, but very strongly on the nature of the impurities along with the plutonium, particularly light elements, because of production of neutrons through (α,n) reactions. Nevertheless, for of Pu holdup in a thick-walled device with a high gamma absorption such as a furnace, assay using neturon detection may be the only practical way to make measurements from the outside.

In their present state of development, the small size of cadmium telluride (solid state diode) gamma detectors,[3] has been a handicap for usual assay applications, but is a boon in a particular type of holdup measurement. Because of their small size (typically they are peckaged in a volume of a few cm^3), highly directional shielding collimators can be fitted around them which do not weigh much, say ~0.5 kg. Such a detector can then be supported at the end of a length of thin wall tubing used as a handle to form a light-weight probe which can be used to trace, at least qualitatively, the deposition of material along otherwise inaccessible pipes, etc.

The difficulty involved in holdup measurements has been an incentive to develop what might be termed "magic black box" approaches to the determination of holdup in a facility. The intent is to create a device such that with a single measurement (or relatively very few), somehow, all the material holdup in a given room of the plant will be measured and accounted for in one fell swoop. In the case of plutonium, the black box has taken the form of a large neutron detector made up of He^3 proportional counters in a flat array embedded in polethylene moderator and sandwiched between sheets of the same material. The response to neutrons is approximately independent of energy.[4] The whole has been appropriately termed a "slab" detector because of its shape. It is too large to be easily moved by hand.

In use it is suspended at one or more points in the room to be assayed, where it samples the neutron flux due to the plutonium. This measurement is supposed to be related to the amount of material giving rise to the flux, through a calibration of the detector, by auxiliary experiments under known laboratory conditions, and by an application of reactor physics type computer codes. Reasons why this method may not work in practice are uncertainties about isotopics, the presence of impuritiss which can produce (α,n) neutrons, the question of how isolated the room being assayed is from neutrons produced in adjoining rooms, the adequacy of the flux sampling scheme and the calculational model, and the fact that by its nature, this kind of measurement does not locate where particular deposits may be, but just gives an integrated effect.

It is however too easy to criticize this approach; what must be kept in mind is that the alternative involves extremely painstaking, laborious, and time consuming efforts to assay every item in the facility. Possibly the importance of this idea is not supplanting an item-by-item search for nuclear material, but to supplement it by an independent assessment which should agree to well within an order of magnitude.

The equivalent black box approach suggested for uranium holdup is a large well collimated NaI photomultiplier gamma detector, the so called "counter telescope".[5] Exposing this to

the relatively penetrating 765 and 1001 keV lines characteristic of U^{238} should in principle allow some correlation with uranium in the holdup. Again, some allowance has to be made for isotopics, absorption, etc. To our knowledge nobody has actually tried this. An attempt in this direction was a 7.5 cm D. x 7.5 cm H NaI photomultiplier detector surrounded with a heavy lead collimator. This in turn was mounted so that it could be pointed at any angle and elevated over a range of about 4 meters. The general intent was to use it as a kind of gamma telescope, i.e. to point it at relatively unaccessible objects or areas and obtain assay values thereby. It was moderately useful, see Figure II.

The general requirements for electronic instrumentation used in holdup measurements have been cited above. Probably the most generally useful combination of qualities for holdup measurements has been a portable instrument which contains, essentially, a detector bias supply, a nuclear pulse amplifier, two channels for pulse height analysis, and a scaler. The original realization of this concept was the Eberline SAM (ESAM) instrument,[6] which has seen wide use, both for holdup and other safeguards applications. The Brookhaven Survey Assay Meter (BSAM),[7] now in its third model version, contains improvements in virtually all the parameters of interest for such portable nuclear instrumentation, together with some altogether new facilities made possible by advances in electronics. It has become the main piece of electronic instrumentation in our measurement of holdup, and indeed, the impetus for its original development were several failings of the ESAM as applied to holdup measurements and other measurements. Figure III shows a BSAM model II with a NaI-p.m. tube detector.

The BSAM can be used with a variety of detectors. For gamma ray measurements where the sensitivity of a NaI-p.m. detector is required, a specially series of all sizes of NaI-p.m. detectors were developed in coordination with the BSAM project. These have, in addition to a ^{241}Am alpha source to furnish a reference pulse, a thermistor which acts to compensate for the different behavior of alpha pulses compared to gamma pulses with temperature change. The results is that gamma ray pulse heights with BSAM are constant over the permissible range of operation of ordinary NaI-p.m. detectors, ~5°C - ~40°C.

The BSAM is also capable of operating with a high resolution detector, such as an intrinsic (hyperpure) germanium detector or CdTe detectors. The hand held germanium detectors were in fact developed in conjunction with BSAM development.

Instruments based on proportional counters may also be operated from the BSAM or the ESAM.

The instrument also incorporates a sophisticated (pocket type) calculator, the HP41C. This furnishes readout, simplifies the process of standardizing the instrument (e.g. arranging for automatic background subtraction), taking data, performing calculations involving data or otherwise related to the work (e.g. attenuation of gamma rays), recording data either in memory or on a miniature auxilliary printer, and reading bar code labels with a light pen, the latter two units being standard available accessories.

The BSAM is constructed so that it can be used slung on the operator's shoulders, with one hand holding the detector, the other hand being free to operate the instrument controls or calculator. For assaying floors, a light weight wheeled cart (nick-named "floor sweeper" from its resemblance to an upright floor vacuum cleaner) has proven convenient. The BSAM chassis is held in a basket mounted on the upright handle at a convenient distance from the operator, while the collimated detector is held a prescribed adjustable distance from the floor it is viewing.

There are now being developed miniature multi-channel analyzers[8] which offer comparable support to detectors as the BSAM, but with the even greater capability for detailed data gathering and analysis capability implicit with an MCA. These have not yet been used in holdup measurements. Such multichannel instruments even though microprocessor based, are more complex and presumably therefore more difficult to operate, particularly under the typically uncomfortable environments of holdup measurements, than the simpler "few channel" instrument such as the BSAM. The comparative value of the small MCA holdup measurement depends on whether furnishing the extra information they are potentially capable of is worth the extra complexity.

Not to be confused with the above possible use of an MCA as a portable survey instrument is its use during hold up measurements in the more usual way as a laboratory bench type of instrument. The MCA used this way is necessary particularly at the beginning of the assay campaign in order to make sure of the isotopics to be encountered in the plant, any unusual backgrounds, etc., and to be available as an arbiter in case anomalies show up in the two channel analysis.

Also found desirable is a small moderately fast two channel oscilloscope available to check the functioning of the electronics, make adjustments, etc.

Nuclear material radiation source standards are a most important part of the assay process. They are essentially the only way of correlating the observed count rate with the amount of material in the holdup at the time of the measurement. The most used of these sources are what are termed "area sources". These are reasonably uniform depositions of UO_2 or PuO_2 fixed on to a sheet of material in some way so as to simulate an area source of radiation, e.g. a glove

box wall covered with a dusting of oxide. Several methods of manufacturing these have been considered or actually tried.[9] The best method seems to be to take transparent plastic with adhesive on one side (a material used to laminate documents) and brush the powdered oxide over the surface. The transparency allows a visual check of the uniformity of disposition. A frame of backing material is left in place surrounding the area from which the backing material has been removed, exposing the adhesive, to which the oxide sticks. The frame backing is then removed and another sheet of plastic is pressed in place so that the adhesive surfaces face one another. Thus an area holding oxide is surrounded by a clean sealed area in which there is no radioactive material. This packaging is rugged enough for uranium; for plutonium, an additional encapsulation is then performed. At appropriate points in the above process the weight is measured on an analytical balance, so that enough information is available at the end to deduce the total amount of nuclear material deposited within the frame. The sheets made have been of several sizes so that particular needs can be met most effectively. The largest have been of the order of 20 cm x 20 cm containing less than 15 g of oxide material. This latter limit is dictated by a desire not to have too much material tied up in any one physically discrete item. It also corresponds, roughly, to the amount of material which the adhesive material in $\sqrt{400 cm^2}$ can hold. More precisely, this depends on grain size. In one instance, the oxide was so finely ground that the adhesive power was used up with far less than the intended load per unit area. It was necessary to get around this problem by mixing the powder with a trace amount of a cellulose-acetone based glue, then coarsely grinding the resultant lumps, and then sieving it to a more appropriate mesh size. Figure IV is a photograph of an area source of the transparent type being made.

Another method of making such area sources which has been tried has been to mix the oxide with silicone rubber. This is poured onto a tray containing a previously cured sheet of silicone rubber free of oxide in such a way that only a prescribed central portion of the oxide free sheet is covered. When the oxide containing silicone rubber has cured, another layer of oxide free rubber is poured over all. In this way the nuclear material, contained in silicone rubber, is encased in pure silicone rubber. This way of making sources is not limited as to the loading per unit area, and is comparatively rugged, but the resulting sheet is inconveniently thick (geometrically) for many applications and has appreciable self absorption.

Discs of uranium foil, about 5 cm in diameter, $\sqrt{0.1}$ mm thick, containing about 4g of $\sqrt{93\%}$ enriched uranium, similarly laminated between transparent plastic sheet, and with the self-absorption factor accurately known have been useful as a gamma check source for uranium assay, and have also served as "point" sources. For the latter to be a reasonable approximation the source needs to be far enough away so that the area of the disc is only a small percentage of the area viewed by the solid angle of the collimator. The amount of material in the disc is sufficient to give an adequate count rate for such distances.

Samples of concrete flooring or of walls are obtained using rotary impact drill motors together with special carbide bits. Using a tool and bit of professional quality, a hole $\sqrt{10}$ cm deep by $\sqrt{1.25}$ cm diameter can be bored in concrete in only 10 seconds with very little effort on the part of the operator. Moreover, nearly all the concrete dust generated is neatly mounded on the floor area immediately around the drill hole. Drilling such sample holes with ordinary rotary drill motors with conventional bits was much more laborious and wore out the drill bits at an unacceptable rate. The rotary impact drill motor speeded up the hole drilling to the extent that the relative slowness with which the drilling dust could be collected using spatulas, etc., was the governing factor determining the rate at which samples could be taken.

This led to the development of an apparatus to pick up the dust quickly and efficiently. It has not yet been used in the field, but shows promise (see Figure V). A small vacuum cleaner whose speed is controlled by a variable auto transformer exhausts air from the top of a small cyclone separator. A suction hose is attached to the input of the separator. The free end of the hose is a $\sqrt{1}$ cm tube which can be inserted into the 1.25 cm diameter bore holes. The tapered funnel bottom of the cyclone separator allows a small sample vial to be attached to receive the solid matter separated form the air stream. The vacuum cleaner exhaust has three absolute cartridge filters in series; a sheet of filter paper in the cyclone separator stretched across the wide portion of the upper funnel poses a barrier against particulate matter that might still remain entrapped in the air stream entering the exit funnel of the separator. The vials can then be capped off and assayed with a NaI well photomultiplier detector. This apparatus was tested for the efficiency with which it collects concrete dust by having it collect a sample of dust, weigh it and then spill it again, followed by another collection weighing, etc. The loss of weight from the successive dust samples averaged less than 1%.

A sheet of lead can be draped over the back of objects being assayed to reduce the influence of radiation from the rest of the plant. About 1/4 inch ($\sqrt{6}$ mm) is a reasonable compromise between portability and isolation. Flexible mats of lead wool covered with plastic of several convenient sizes are available commercially and are more convenient albeit offer less attenuation. Since they are appreciably more expensive than plane sheet lead, there is however reluctance to use them in an application where they are likely to become contaminated.

As an example of the use of a lead shield, consider an assay of holdup on an interior wall of a glove box. One way would be to pass the sheet of lead into the glove box and prop it up against the wall surface being assayed. The detector is then used to view outside of the box. A correction would then have to be applied for attenuation through the glovebox wall. An alternate way of assay would be to insert the radiation detector into a rubber glove and in that way allow the detector to view the inside surface of the glove box directly, avoiding the uncertainty of the correction factor. The lead sheet could then be outside the glove box. The glove should of course be free of radioactive contamination, which means that it probably should be a freshly applied one. Simple sketches and polaroid photos of work areas, ductwork, glove box interiors, etc., prove useful as memory aids during the data reduction process.

Assay of concrete plant flooring

The floor is divided by a convenient sized rectangular grid (a good method is a dab of spray paint at each intersection of chalk lines), each denoted by a double subscript. The size of each box should be dictated by a preliminary survey to balance the detail needed for a fair representation of the distribution of holdup, against the total number of grid boxes that would have to be sampled.

Each grid box is assayed at several interior locations according to a prearranged pattern using e.g. a BSAM gamma detector, mounted in the "floor sweeper". The apparatus is calibrated very simply by placing it over one of the area standards on a clean stretch of floor. The overall count is assigned to that box. The results are ranked and grouped, and then some are selected from each group for sampling of the concrete. This is done using rotary-impact drills, the required base depth being determined by a prior study of the incremental depth versus radioactivity. Complete recovery of the drill dust is attempted; it is packaged in a vial, weighed in a simple chemical balance, counted with a NaI-well crystal and sent to NBL for a chemical and isotopic assay. The correlation between the surface count assay seen with the floor sweeper, and the actual chemical-isotopic assay when it becomes available, furnishes a calibration for the whole floor area. In the meantime, the well counter assay furnishes a working correlation.

That there should be a correlation between a surface gamma measurement and the material recovered from the bore is somewhat debatesble. A simple picture of how special nuclear material works into a concrete floor is that the porous concrete acts like a chromatographic column, with repeated spills of presumably similar solutions just causing deeper migration into the concrete, with the concentration profile retaining the same form. Radioactivity versus depth studies confirm that this picture is valid some times. Often however this simple picture even if true initially, is disrupted by various activities during the plant history, such as pouring of a new floor over the old, chipping out the old surface, with subsequent resurfacing, sealing the old floor with a liquid barrier paint, etc. In case the correlation of surface radioactivity with that of the powdered core sample using the NaI well counter is poor, then the core sampling has to be done more extensively.

Assay of duct work and piping

Measurements are made with gamma detectors, typically the BSAM system. The measurement points are marked and numbered with felt tip pens and coordinated with plant furnished engineering drawings of the duct work and piping. Measurements are made at intervals depending on how much material seems to be involved, and the apparent gradient of the deposition. Attempts are made to shield the section being assayed from radiation from other objects by the use of sheet lead shields. Uncertainties in how to calculate attenuation are compounded in the case pipes and ducts by difficulties in allowing for geometrical effects. What has been found effective here is to obtain from the facility sample of the pipe or duct, line the inside with an appropriate area standard, and use this to calibrate the detector-collimator. The area source has to fit inside without overlap and present within the solid angle viewed by the detector an appearance of a uniform interior coating. The actual measurement is taken for the real object, if possible, from the top, bottom, and two opposed horizontal directions, and an average taken which is compared with the calibration to establish the local density of deposited material at the measurement point. This method is favored since the actual measurement geometry can be duplicated accurately, eliminating a potential source of error. Holdup measurements based on such calibrations have agreed quite well with the amount of material subsequently recovered. However, in some cases, attenuation calculations based on the known or assumed wall thickness have been used, particularly when clean samples were not available. In any case the assay results are expressed as the apparent amount of material deposited per unit length, and noted on a simple dimensional sketch of the system made as the assay procedes. The assumption that a measurement is representative of the immediately surrounding lengths of duct or pipe allows an estimate of the total holdup in the particular duct or pipe system to be made.

Walls and ceilings

A preliminary survey of walls is made to find those areas where liquids, etc. from process equipment might have soaked in. Such regions, if found, are assayed the same way as are floors, i.e., core drillings are made, etc. Wall areas where there is not much material are assayed using a collimated detector calibrated for viewing areas (the so-called "counter

telescope"). Figure V shows a counter telescope developed for such measurements. The smaller SSAM detector and electronics has also been used in a telescope mode.

There are instances where there was enough material on the walls so that profiles of the deposition of nuclear material as a function of height above the floor were of interest. The concentrations typically decrease with height above the floor, sometimes there is a maximum at, say, 50-100 cm height.

Often both sides of a wall have surface depositions of nuclear material, with the wall having insufficient attenuation to allow each side to be assayed in isolation of the other. An approach to measuring holdup in this case is to make an area measurement from one side of the wall, with a lead sheet on the far side to isolate the wall from the rest of the adjacent room, and then repeat this process for the other side. If the respective count rates are $C_{1,2}$, the area calibration factor is k, the attenuation through the wall is $0 \leq a < 1$ and the mass of nuclear material per unit area deposited on the respective sides is $m_{1,2}$, then

$$c_1 = km_1 + kam_2,\ c_2 = kam_1 + km_2$$

which has the solution

$m_1 = (c_1 - ac_2)/k(1-a)$, $m_2 = (c_2 - ac_1)/k(1-a)$ so long as $a \neq 1$, i.e., there is some attenuation. The attenuation can be measured by using a source strong enough to overpower the deposited material. This source is assayed through the wall and then an equal distance away without the wall intervening, and the two assays compared.

Ceilings seem generally to have low fairly uniform depositions and have been assayed using the counter telescope approach.

Glove boxes and furnaces

As a preparation for assaying glove box walls or other contents, it is useful to have at least some of the gloves replaced or to have them removed altogether and the ports blanked off with clean (free of contamination) sheets of thin, low z, material such as lucite. These can then be used as "windows" through which a collimated detector can be aimed at objects inside the glove box. Lead masking sheets, as suggested previously are quite useful for glove box assay.

Objects within the glove boxes are idealized as points, line, or plane (area) sources. The detector's response to a point source is calibrated using a well characterized fuel pellet or metal disc source at a known distance. The response at other distances of an actual object is allowed for using an inverse square of the distance law. A line source of radiation could be calibrated for by placing a point standard at successive positions and assaying it along a line perpendicular to the detector axis. An actual line source at some other distance can be allowed for using the fact that the intensity of radiation from a line source should vary as the inverse distance. In practice, however, such objects have been treated where possible as duct work and pipes.

Large, thick walled, furnaces are a problem because of the uncertainty involved in correcting for the absorption. In one instance, the detector was passed into the furnace on the conveyor system at the end of a long cable to the supporting electronics in order to get an unimpeded view of the insides.

Recommendations concerning plant construction

Many of these can be inferred from the above text. The basic point to be made is that process lines should be constructed with a view towards minimizing the creation of holdup and facilitating its measurement and removal. As examples of the former: Duct work should be designed and layed out with some consideration for the gas dynamics involved. Process areas where wet chemistry takes place or liquids are handled should be constructed as far as possible with trays to catch the inevitable leaks. Floors should be sealed before use. Square tiling should be avoided in favor of seamless covering.

As examples of the latter: Duct work should be demountable. Furnaces (or similarly thick walled objects) should be provided with radiation probe tubes allowing insertion or permanent placement of radiation monitoring devices.

In conclusion

Holdup measurements combine features of detective work, a treasure hunt, and a lot of detailed measurements performed under often unpleasant circumstances.

Most recent holdup measurements has been prompted by plant decommissioning. However, the basic qualities of special nuclear material that make it the object of search and recovery efforts will not likely change: Since the material poses a health hazard, generally has a high intrinsic value, and must be safeguarded against the ever present threat of diversion, holdup measurements will be a necessary part of even a viable nuclear manufacturing industry.

Properly designed and maintained manufacturing facilities would decrease the burden of such measurements.

Acknowledgements

We would like to acknowledge in particular our sometime co-workers and companions in holdup measurement, in particular, D.M. Gordon of BNL (TSO) and J.W. Tape of LASL (Q1), for their contributions to this art.

While the work reported here was done under U.S. N.R.C. auspices, the authors would not wish to imply that their methods, opinions, etc., have any official sanction. In fact it is our intent that this article furnish points for discussion rather than be considered definitive.

References

1. This work was done in conjunction with ORTEC (Oak Ridge, Tennessee), and separately, PGT (Princeton Junction, N.J.). Both these companies now offer such detectors as a regular commercial item.

2. Manufactured by the Eberline Instrument Company (Santa Fe, N.M.).

3. Units we used were from the EMD Corp. (Watertown, MA.)

4. "Total Room Holdup of Plutonium Measured with a Large-Area Neutron Detector", J.W. Tape, D.A. Close, R.B. Walton, Proceedings INMM 17th Meeting, p. 533, June 22-24, 1976.

5. Possibly originally a suggestion of W. Higinbotham and co-workers at BNL.

6. Still offered by Eberline Inst. Co.

7. "A New Survey-Assay Meter for Portable Applications", M.S. Zucker, et. al. Proceedings 1st Annual ESARDA Symposium, p. 289, April 1979. A commercial version may be available from IRT (San Diego, CA.).

8. The smallest to date was developed by J. Umbarger and co-workers at LASL.

9. The particular ones we have used were developed by A. Gody, U.S. N.R.C. Region I (King of Prussia, PA.) in conjunction with M. Degen and co-workers at BNL.

Figures

1. Count rate from disc source as it is moved on a plane perpendicular to the detector-collimator axis on a radius passing through the point of intersection of the axis and the plane.

2. A counter telescope, consisting of a heavily collimated large sized NaI-photomultiplier which can be pointed in any direction and raised over a range of about 4 meters (CN 8-594-78).

3. The BSAM model II used in much of this work is here shown with a small collimated NaI photomultiplier detector designed to be used with it. BSAM model III units are now being produced (CN 1-1274-79).

4. A planar (area) source of the type using transparent adhesive backed film, after manufacture in a glove box (CN 9-513-76).

5. A prototype apparatus developed at BNL for safely and efficiently collecting radioactive dust, as from a concrete bore hole (CN 5-56-81).

Figure 2

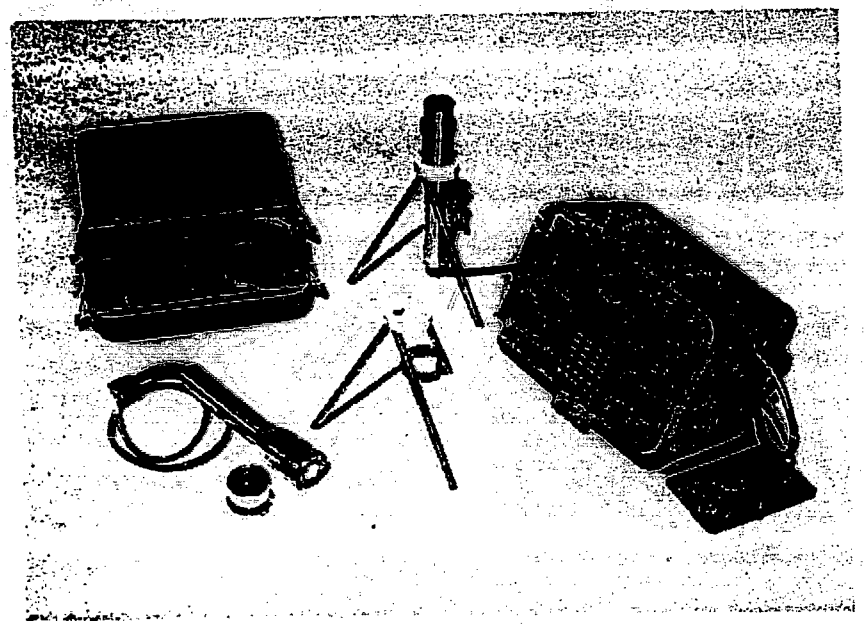


Figure 3

Figure 4

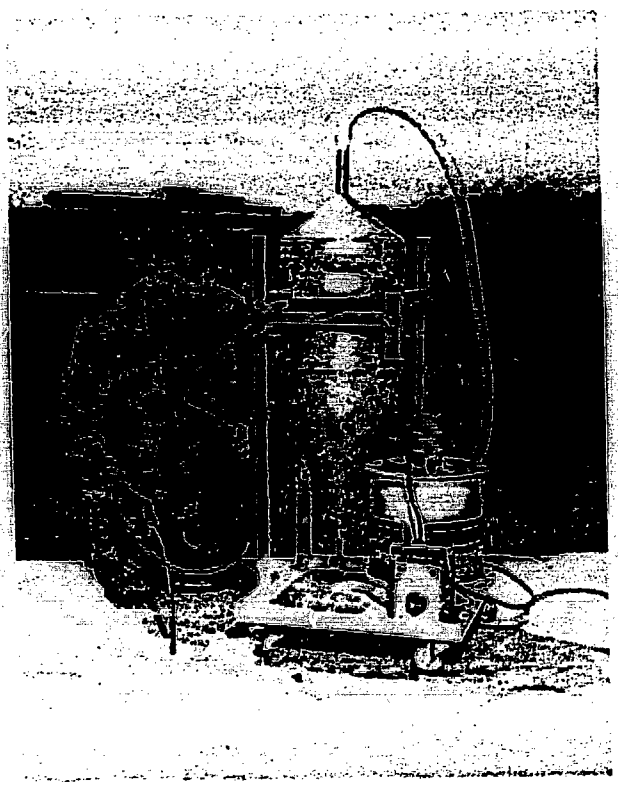


Figure 5