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TITLE: LEARNING TO LIVE WITH HOLDUP

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LEARNING TO LIVE WITH HOLDUP*

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ABSTRACT

Holdup of special nuclear materials in processing facilities is recognized by facility operators and regulatory agencies as an insidious materials control and accounting problem. However, there have been few serious efforts to address holdup as a materials accounting problem and to accommodate the legitimate concerns of both groups. This paper reviews past efforts and identifies several key elements relevant to resolving the problem in a pragmatic fashion. These key elements relate to the recognition of holdup as a serious materials accounting problem, innovations in holdup monitoring and their limitations, the role of modeling and sampling in holdup estimation, and the potential value of plant-specific materials accountability requirements. Suggestions are offered for developing cost-effective procedures for holdup measurements/estimation, combining available technologies with properly designed sampling plans.

I. INTRODUCTION

Holdup of special nuclear materials (SNM) is defined as the material remaining as residual inventories in process equipment after the runout of bulk material. Thus, holdup is different from in-process inventory. It is a hidden inventory, distributed over extensive surface areas and in numerous process equipment. The fact that it is "hidden" lends itself to interpretation as unidentified loss and diversions. Since unidentified losses or diversion are serious safeguards concerns, holdup is a problem that needs to be addressed during all phases of plant design, safeguards system design, construction, process development, and process safety assurance.

There are numerous examples of reported large inventory differences (IDs) at DOE facilities and NRC-licensed facilities. Different interpretations have been given to those IDs by facility operators ("holdup is a fact of life"), regulators ("how should these numbers be properly explained?"), and the media ("how many bombs are missing?"). These large IDs often have been subsequently proven to be caused by holdup. Emerging stringent demands on special nuclear materials accounting have created a new awareness about the problems of holdup--new, that is, to some process engineers, administrators, and regulators. It is desirable for the entire nuclear materials production and safeguards community to learn about holdup and to live with it, because these problems cannot be wished or legislated away.

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II. MECHANISMS OF HOLDUP

Holdup is not a new phenomenon. Materials generally accumulate in cracks, pores, and zones of poor circulation within and around process equipment. Some processes lead to the accumulation of sizable and sometimes continuously increasing amounts of materials in difficult-to-recover form. The walls of process vessels, plumbing, ductwork, glove boxes, and filters become coated with SNM during processing. In addition, SNM and its often corrosive chemical environment may interact with the components of the equipment and become tenaciously adhered to the surfaces, causing another form of residual holdup. There are other mechanisms of material accumulation in process vessels, including:

- unplanned buildup in pipes and ducts due to poor layout and construction features;
- electrostatic deposition of particulates;
- post precipitation due to delayed reactions; and
- coagulation and sedimentation in storage tanks.

From a detailed knowledge of process chemistry, behavior of material forms within a plant, and plant layout, it is possible to reasonably predict the locations of holdup deposits during normal operations. However, depending on frequencies of process upsets, quality of facility maintenance, operational skills of the personnel, cleanout practices, and plant throughput, the magnitude of holdup can vary drastically from one plant to another using the same process.

III. STATUS OF HOLDUP ESTIMATION

The role of holdup as a problem in nuclear material safeguards was recognized very early in the US.[1] For holdup measurements, in situ assay techniques are preferable to process-disruptive and time-consuming cleanout measurements. The general principles of these nondestructive radiation measurement techniques are well understood, and their applications to safeguards measurements are described in detail in several publications prepared for the safeguards community.[2,3] Assay procedures acceptable to regulatory staff are detailed in regulatory guides for the measurement of uranium and plutonium.[4,5] Accuracies in holdup measurements are generally poor[6,7] because of complexities of the residual deposition patterns and the attendant calibration problems for nondestructive assay (NDA) measurements. Other factors that contribute to poor measurements include high background radiation levels, difficult-to-access regions of plants, and equipment where there is holdup. There have been many suggestions to avoid obvious bias in standards and to develop facility-specific calibration procedures.[8-9]

Historical data gathered during inventories are often considered as a useful source of information. However, large measurement errors and incomplete recording of many relevant factors make this source of information of limited value to statistical model development. The pressures to quickly explain IDs and disincentives in publishing IDs in the open literature further contribute to the limited value of this source.

The difficulties associated with the estimation of process holdup are reflected in proposals to use secondary methods of measurement.[10-14] Design considerations to minimize holdup have been published in a regulatory guide[15] to meet safety requirements and to ease holdup estimation problems. In the past, there have been attempts to develop estimates of the contents of process vessels with the help of elaborate computer programs and previous inventory measurements, operating data, and on-line process measurements.[16-18] These efforts, still in the early stages of development, are intended to be specific to unit operations.

From experience gained over the past decade in materials accounting, it can be generally stated that

(1) NDA techniques are the only way of measuring holdup on a cost-effective basis;

(2) it is not possible now nor will it be possible in the near future to measure holdup in most equipment with an error of less then 10% (or in many cases <20-50%);

(3) at most facilities, holdup is dispersed over many square miles of surface areas, so that available resources preclude measuring more than a small fraction of all holdup;

(4) the large measurement and sampling uncertainties in (2) and (3) imply that when one estimate of facility holdup is subtracted from another for purposes of estimating the change in holdup over an inventory period, the resulting error is quite large; and

(5) during startup with clean equipment, holdup almost always increases (as something of a plating-out process takes place) and because material is lost to the accounting system, IDs behave accordingly.

Several of these recognized limitations are among the reasons why present regulatory policies do not allow in-place measurements of holdup to be incorporated into the ID. As a consequence, changes in holdup over several inventory periods influence both the average ID and the period-to-period fluctuation around that average, and propagation of measurement errors alone often does not adequately explain ID behavior.

IV. HOLDUP MEASUREMENTS AT PROCESSING FACILITIES

Holdup is characterized by the materials that are difficult to locate, sample, identify, analyze, and quantify. In materials accounting, holdup is often ignored or improperly measured. Thus, holdup has adverse effects on the quality of physical inventories and on materials accounting programs. As part of an investigation for the Nuclear Regulatory Commission, Los Alamos[19] evaluated several types of holdup, including holdup of uranium and plutonium at three processing facilities. The various equipment involved in these holdup measurements were

(1) HEPA filters at a plutonium processing facility,

(2) several air filters and batch calciners, and a continuous precipitator and a rotary drum filter at the uranium scrap recovery facility, and

(3) several air ducts at a HTGR fuel fabrication facility.

Measurements were performed using shielded and collimated NaI(Tl) detector(s) and dedicated multichannel analyzer system(s). Calibration standards for the detector system(s) were fabricated to resemble the components being measured using known amounts of PuO_2 or UO_2 dispersed on suitable media. Transmission and attenuation corrections were determined using thin sources of PuO_2 or UO_2.

Confirmatory measurements for plutonium were performed using a neutron coincidence counter. For uranium, confirmatory measurements were done on cleanout residues in favorable geometries using high efficiency gamma-ray spectrometry.

V. CONTROLLED STUDIES OF SNM HOLDUP

A recent comprehensive report[19] on holdup estimation contains details of controlled experiments designed to measure uranium holdup in several unit processes common to nuclear reactor fuel conversion/fabrication lines. Three of these unit processes were

- a dust generating operation at a HEU processing facility,
- an ammonium diuranate (ADU) precipitation and calcination process, and
- a solution loop system circulating uranyl solutions.

Total throughput of uranium through these experimental facilities ranged from 50 kg to about 100 tons. The quality of measured holdup data during these controlled experiments was improved by at least an order of magnitude through the use of carefully selected radioactive tracers[14] and specially designed calibration standards. Tracers, at concentration levels of about one part-per-billion, were homogeneously incorporated into the process materials. Considerable attention was paid to fabricate calibration standards compatible with the equipment measured and the distribution of holdup therein. This also contributed to improve the quality of holdup data from noninvasive, nondestructive assays by gamma-ray spectrometry using NaI(Tl) detectors. Some of the general features of the controlled experiments are summarized in Table I.

These controlled studies have demonstrated that well-designed experiments at large facilities combined with reliable measurements can be used to develop holdup estimation models. The quality of the holdup data being the key to the successful model development, it is important to invest sufficient effort to minimize measurement uncertainties. Data from these controlled experiments were extremely satisfactory: accuracy of holdup measurements were within 3-15% of the "cleanout" values, and a majority of these measurements showed precisions of less than ±5%. Thus, the controlled studies were useful in developing mathematical models of holdup in each of these equipment. Details of the modeling approaches, their advantages, and their limitations are summarized in the following section.

VI. MODELING AND SAMPLING FOR HOLDUP ESTIMATION

Like many physical processes, the accumulation of holdup is amenable to modeling. Frequently, holdup behaves as a smooth function of time (e.g., increased deposition on a filter serving a steady-state operation) or of space (e.g., holdup at one section along a length of piping may be very similar to holdup at an adjacent section). Models that effectively characterize "global" behavior lead to improved estimation. That is, holdup estimates based on measured values and the model are superior to "local" estimates based on a measured value alone. The phenomenon is not peculiar to holdup estimation but is common to most statistical applications of modeling.

Despite its value in many situations, modeling has limitations that preclude its universal use. If operation-specific models are required for numerous modes of operation or if material accumulation over long periods of time must be

TABLE I

EXPERIMENTAL PARAMETERS OF CONTROLLED HOLDUP STUDIES

Controlled Experiments	Material Forms Involved	Tracer Used	Experimental Parameters Varied	Equipment Involved in Holdup Measurements
U-dust generation	U_3O_8 powder, and incinerator ash containing U_3O_8	^{95}Zr-Nb (Neutron-activated U_3O_8)	40-200 µm particle size, air flow rates from 5 to 100 cfm	Glove box, ducts, elbows, tee, prefilter
ADU-precipitation and calcination	$UO_2(NO_3)_2$, ADU, and U_3O_8	^{46}Sc as Sc^{3+}	pH of solutions, mixing rates, calcination temperature	Dissolver, precipitation filters, calciner trays, calciner
U-Solution loop	$UO_2(NO_3)_2$ and UO_2F_2	^{46}Sc as Sc^{3+} and $(ScF_6)^{3-}$	Flow rates, materials of construction, pipe dimensions	Pump, pipes, elbows, tees, unions, valves, terminal valves

characterized, then conducting the controlled experiments can become expensive. Also, models developed using poor quality data are often of limited worth. Thus, modeling should be approached judiciously--being a tool implemented for high priority equipment and when there is a good chance of success.

The use of models to obtain holdup estimates has been discussed in the literature[19-20] and are not elaborated here. Supposing that estimates of holdup at individual locations are obtainable (whether based on models or not), a larger issue involves efficient use of available resources. As noted in Sec. III, measuring <u>all</u> holdup is impossible at most large facilities. In order to acquire a facility-wide understanding of holdup, sampling procedures are a necessity. Related examples of sampling reported in the literature on international safeguards work include papers on such subjects as attributes/variables procedures and MUF-D statistics. Other applications include bulk sampling for analytical measurements and the DOE-recognized sampling plans for bimonthly inventory work for stored items of SNM.

Of interest is the development of sampling design that is methodologically correct and cost effective. The sources of information used for sampling design include

(1) a complete listing of equipment containing holdup at the facility,

(2) anticipated nominal amounts of holdup for each item in (1) and anticipated variation in those nominal amounts,

(3) measurement uncertainties associated with the holdup measurements to be made, and

(4) budgetary data to compute costs.

Once this information is available, the development of an optimal sampling plan for holdup is equivalent to solving an optimization problem.

Before discussing that optimization, it is useful to briefly review the above sources of information. The first source, an equipment listing, can usually be obtained from facility blueprints. An exhaustive facility description defines the "population" from which the sample of items to be measured is drawn.

Given the listing in (1), anticipated nominal amounts of holdup are needed for the items in the population. These amounts may be somewhat crude approximations. Effective sampling designs put most effort, all other things equal, into strata of items where the most holdup is expected. Moreover, holdup in a relatively homogeneous stratum--i.e., in a type of equipment with roughly constant holdup from item to item--is estimated more accurately than holdup in a nonhomogeneous stratum; this behavior is similar to that of errors in analytical measurements based on bulk sampling of homogeneous and nonhomogeneous materials.

Measurement uncertainties are necessary for error propagation. A measurement control program using good holdup standards is useful here, as are comparisons of cleanout data to previous NDA measurements.

Budgetary data are valuable for many reasons. From a technical standpoint, it is not possible to design cost-effective holdup plans without knowing what the costs are. From a management standpoint, cost figures are useful in allocation of resources for holdup monitoring relative to allocation for other accounting activities. In calculating costs, a variety of factors must be considered, including lost production time, "direct" costs of making the measurements, and data analysis/reporting.

For any specified sampling plan, aforementioned information in (1)-(4) can be used to compute the cost of that plan and the standard deviation of its associated holdup estimate. Space here prohibits a detailed development of the calculation of the standard deviation using the measurement uncertainties in (3) and the within stratum variabilities in (2); that calculation is described elsewhere[21] and related methodology can be found for other applications.[22,23]

Somewhat typical results for a single stratum are displayed in Fig. 1, where holdup in 15 furnaces in a calcination line is estimated (see Ref. 21 for details). Adopting the most expensive sampling plan, which measured all furnaces, leads to a standard deviation of ~40 g, a limit determined by measurement sensitivity. A diminishing returns shape, displaying the roles of less expensive plans, is apparent in Fig. 1. That is, once an adequate number of randomly sampled furnaces are measured, there is little benefit gained from the (expensive) process of measuring the rest.

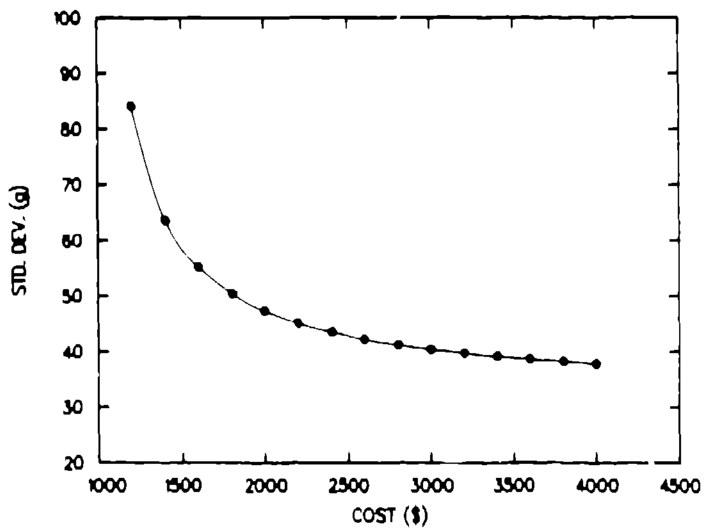

Fig. 1
Relationship between standard deviation and cost of holdup measurement (Ref. 21).

Generally speaking, a computer subroutine that takes a specified, facility-wide sampling design and computes its cost and its associated standard deviation can be imbedded in an optimization code, leading to solutions of important problems. One such problem concerns finding the plan, of all possible plans costing less than some prescribed amount, that provides the best holdup estimation. In this fashion, formal selection of the desired sampling plan is achieved.

VII. DISCUSSION AND CONCLUSIONS

Holdup measurements in large processing facilities are difficult and will remain so because of the complexity of the problem itself and inherent limitations of NDA techniques. One of the alternatives to NDA measurements is "cleanout" measurements using destructive and nondestructive analyses. This alternative is not only time-consuming, expensive, and process disruptive, but it cannot provide timely safeguards information. An inevitable reality at most processing facilities is the difficulty of assigning high priority for holdup measurements relative to other concerns. It is, however, important for plant operators to recognize that a knowledge of holdup problems can make valuable contributions to plant safety, process efficiency, and nuclear material safeguards.

Discussions of key elements of holdup as a materials accounting problem (and a potential safety problem) demonstrate that it is important to recognize that a practical approach be developed to satisfactorily deal with this issue without seriously affecting the primary missions of production facilities. Some approaches to dealing with holdup as a safeguards issue include the following.

(1) Several design features of process equipment and plant layout could help minimize and localize the problems of holdup. Narrow, tall holding tanks (instead of Raschig ring filled tanks), unitized glove boxes, drain lines that allow both draining and flushing of select regions of pipe lines, etc. are features that can be successfully incorporated in the design of new and/or refurbished plants. For new plants, it is possible to gather good holdup data during the pilot-scale and full-scale operations. Such data can be used to develop holdup models for major process equipment.

(2) For older plants, efforts should be made to systematically gather holdup data during shutdown by <u>in-situ</u> measurements followed by cleanout measurements. This would help to develop proper calibration factors for NDA techniques to estimate residual inventories when cleanout measurements are not practical.

(3) It is important for operators of processing facilities to develop information on process upset conditions and resulting impacts on holdup. This would allow for planned shutdown of the facility for cleanout before inventory if there had been process disorders that would impact holdup and therefore the ID for that inventory period.

(4) Advances in radiation measurements, such as high resolution detectors and portable analyzers, have led to NDA techniques readily adaptable to holdup measurements. Specially designed instruments and calibration standards for holdup measurement, complimented with a measurement control program, can greatly improve the reliability of holdup measurements.

(5) Instead of trying to establish a common yard stick to evaluate the materials accounting systems of diverse plants, it would be prudent to develop safeguards systems that are unique to each facility and attempt to make each of them cost-effective. It is also desirable to design plant-specific procedures to assure the reliability of holdup estimation.

(6) Because variations in holdup between inventory periods are more important to IDs than are the actual magnitudes of holdup, it is highly desirable to minimize process variations that could cause major changes in holdup levels.

(7) Measuring holdup in all pieces of equipment is not a viable option in terms of time, cost, and radiation exposure to personnel. A 100% sampling to measure holdup is not only impractical but unnecessary from a materials accounting point of view. Central to holdup monitoring is the concept of sampling, the principles of which are well known and widely understood. Sampling concepts can be used to design an efficient, cost-effective plan for in-place holdup monitoring.

Current regulatory practices to prevent diversions of SNM are based on the calculation of inventory differences and the limits of error of these inventory differences (LEIDs). Reliable measurements and estimates of inventories are essential to this regulatory process. Statistical estimation models, combined with a carefully developed sampling plan, have potentials to assist plant operators in improving the overall quality of holdup estimation as part of periodic inventory development. However, the development of useful estimation models hinges on the quality of data and the stability of process operations. In developing and using estimation models, it is important to recognize their limitations and use this approach judiciously.

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