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ESTIMATION METHODS FOR SPECIAL NUCLEAR MATERIALS HOLDUP*

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ABSTRACT

The potential value of statistical models for the estimation of residual inventories of special nuclear materials was examined using holdup data from processing facilities and through controlled experiments. Although the measurement of hidden inventories of special nuclear materials in large facilities is a challenging task, reliable estimates of these inventories can be developed through a combination of good measurements and the use of statistical models.

I. INTRODUCTION

One of the basic elements of a system for materials safeguards is materials accountability, which includes measurement, accounting, and other procedures designed to provide an accurate knowledge of the quantities and disposition of materials. Section 70.51 of Title 10 of the Code of Federal Regulations requires, in part, that certain licensees of special nuclear materials (SNM) conduct at specified intervals physical inventories of SNM in their possession under the license. The accumulation of SNM in process equipment as hidden inventories in the form of residual holdup following shutdown, draindown, and cleanout generally has adverse effects on the quality of physical inventories and on materials control programs. Residual holdup is characterized by the materials that are difficult to locate, sample, identify, analyze, and quantify. The residual holdup of SNM may be defined as the inventory component remaining in and about process equipment and handling areas after those collection areas have been prepared for inventory. Regulatory Guides 5.37 and 5.23 provide guidance for the assay of residual uranium[1] and plutonium[2] in processing facilities.

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, continually increasing amounts of SNM in difficult-to-recover form. The interior surfaces of process vessels, plumbing, ductwork, glove boxes, and filters often become coated with SNM during materials processing. In addition, SNM may chemically interact with the components of the process equipment, causing another form of holdup. The amount of SNM in residual holdup must be small for efficient processing and for hazards control. In practice, however, the total amount of SNM holdup is significant relative to plant inventory differences. This points to the need for better design of processing facilities and improved methods of holdup estimation.

As a result of the stringent requirements for the timely detection of the losses of SNM and in recognition of the difficulties of measuring holdup, the US Nuclear Regulatory Commission sponsored a research study at Los Alamos National Laboratory. The primary objective of this investigation was to explore the possibilities of developing statistical estimation models for the holdup of SNM at processing facilities. The task of gathering holdup information and the development of holdup estimators for specific processes underwent several stages of examination. Historical data available from highly enriched uranium (HEU) processing facilities, which were gathered as part of periodic inventory development, were first considered as a readily available source of long-term holdup data. Unfortunately, the poor quality of these data made this source of information of limited value to statistical model development. The next step in gathering good quality holdup data was through carefully designed measurements of SNM holdup at two of the materials processing facilities of the Los Alamos National Laboratory. Selected measurements conducted over a period of one year showed that certain equipment, such as air filters and calciners, lent themselves to good quality holdup measurements and model development. The value of these models was further confirmed when controlled experiments were performed involving high quality data collection using radioactive tracers. Complete details of the measurement methods used during this investigation and the modeling approaches are contained in the final project report submitted to the NRC.[3]

In the following sections, we summarize a few of the controlled experiments and process facility

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measurements carried out during this investigation. Mathematical models are provided to illustrate the different approaches used in developing estimation models of residual holdup.

II. CONTROLLED EXPERIMENTS

The controlled experiments were designed to measure uranium holdup accumulated during dust generating operations of fuel fabrication, feed dissolution processes, ammonium diuranate (ADU) precipitation and calcination, pulse column operation, and the circulation of uranyl solutions through pipes and pipe fittings. Total throughput of uranium in these experimental facilities ranged from 50 kg to about 50 tonnes. 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 and specially designed calibration standards.[4] Tracers, at concentration levels of about one part-per-billion, were homogeneously incorporated into the process materials. Considerable attention was paid during these experiments to fabricate instrument calibration standards compatible with the equipment measured and the distribution of holdup therein. This improved the quality of holdup data from noninvasive, nondestructive assays using gamma-ray spectrometry.

Four unit processes chosen for controlled experimental study were
(1) an ADU precipitation and calcination process,
(2) a dust generating operation at a HEU processing facility,
(3) a liquid-liquid extraction system, and
(4) a solution loop system circulating uranyl solutions.

Complete descriptions of these experimental facilities and detailed discussions of results are presented in Ref. 3.

Because of space limitations, only the experimental study of holdup from ADU precipitation and calcination is considered in detail here. We simulated the generic process involved in ADU precipitation and calcination and measured the holdup of uranium in a dissolver, a batch precipitator column, several filters, a calciner, and several calciner trays.

During this experiment, ^{46}Sc was used as a tracer to measure the residual amounts of uranium in the processing equipment using noninvasive gamma spectrometry. The precipitation column used for this experiment was a stainless steel cylinder, 20 cm in diameter and 1 m in height. Associated equipment used for precipitation of ADU is shown schematically in Fig. 1.

U_3O_8 was used as the starting material for this experiment. Each batch contained 1 kg of uranium, which was dissolved in nitric acid, and to this solution ^{46}Sc tracer ($\sim 10^8$ Bq) was added as Sc^{3+}. The uranium nitrate solution was vacuum-transferred to the precipitation column and precipitated as ADU using NH_4OH, while the

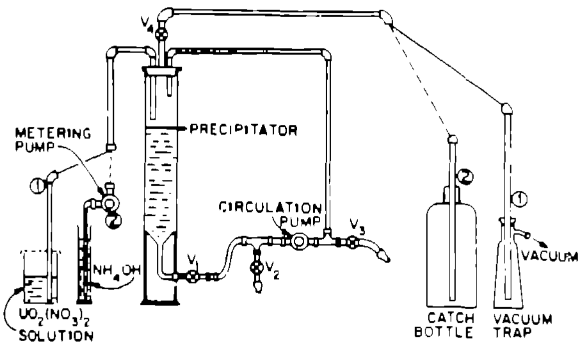

Fig. 1.
An isometric view of the ADU precipitation column: ① solution transfer, ② ammonia addition.

contents of the precipitation column were agitated vigorously by a circulation pump. The ADU was filtered using large Buchner filters, and the ADU cake was calcined in a Lindberg furnace. This process was repeated until a total throughput of 52 kg of uranium through the experimental system was obtained.

After each batch processing, the uranium holdup in the dissolver, precipitator column, filters, calciner trays, and the calciner were measured nondestructively using a specially mounted NaI(Tl) detector-based gamma spectrometry system. Several cleanout measurements were also performed during this series of experiments to confirm the NDA measurements of holdup. The NDA measurement data were used to develop holdup models for the various pieces of equipment used in this experimental study.

The NDA measurement of the holdup of uranium in the precipitator column offered more challenges than the other equipment used in this experiment. The residual ADU in this apparatus was not uniformly distributed, although the profile of this distribution remained more or less the same while the experimental conditions were not altered. The holdup profile of ADU in the column was periodically monitored using a small, essentially unshielded NaI(Tl) detector setup ("Samson," manufactured by Eberline Instruments Co.) to count the high-energy gamma rays from the ^{46}Sc tracer used in these experiments. The spatial resolution of the detector was about 6 cm FWHM for the column geometry. The length of the precipitator column was divided into 16 equal segments, and 17 measurements were made at the boundaries of these segments. Details of these profiles are further discussed in Section IV.

III. HOLDUP MEASUREMENTS AT PROCESSING FACILITIES

As part of this investigation, holdup measurements were conducted at three processing facilities. The various pieces of equipment involved in these holdup measurements were
(1) high-efficiency particulate air (HEPA) filters at the plutonium processing facilities of Los Alamos;

(2) several air filters and batch calciners, a continuous precipitator, and a rotary drum filter at the uranium scrap recovery facility at Los Alamos; and
(3) several air ducts at the HTGR fuel fabrication facilities of General Atomic (GA) Technologies, Inc.

Again, because of space limitations we present only the details of the HEPA filter measurements.

The holdup measurements of plutonium on HEPA filters were performed using a shielded and collimated NaI(Tl) detector installed on top of a glove box about 18 cm from the HEPA filter. A multichannel analyzer system was used to scan the gamma spectrum, and the 320-470 keV region was integrated to estimate the holdup on this HEPA filter. Calibration standards for this detector system were fabricated to resemble the filter being measured, using known amounts of PuO_2 dispersed on a similar filter medium. Transmission and attenuation corrections were determined using a thin source of PuO_2.

Confirmatory measurements were performed on the filters at the end of the experiment period using a neutron coincidence counter to determine the plutonium content. The coincidence counter measurement was within 8% of the in-place NDA estimates of the holdup of plutonium.

IV. MODELING APPROACHES

A. Introduction

Like many physical processes, the accumulation of holdup is amenable to modeling. When facility operation is stable, the holdup in a piece of equipment behaves as a smooth function of time, perhaps gradually increasing or remaining (nominally) constant. This aspect of "temporal continuity" in holdup behavior can often be captured through modeling. A "spatial continuity" may exist as well. For example, holdup at a particular location may be very similar to that at locations nearby. Proper combination of all such relevant information (formalized through use of a model) leads to holdup estimation much improved over reliance on a single measurement value. A more lengthy discussion of holdup modeling is given in Ref. 5.

In succeeding sections, results are discussed from several controlled experiments where holdup was carefully studied. These experiments served to illustrate a variety of points, including when modeling is useful and when it is not. Also, the benefits and limitations of modeling in a number of circumstances became apparent.

B. Modeling With Respect to Time: Increasing Holdup

Consider data obtained from four air filters and displayed in Figs. 2 and 3. Holdup on filters, like holdup on many other pieces of equipment, undergoes something of a life cycle. The initial conditions of little or no holdup are followed by a gradual accumulation of material. Finally, the filter is replaced (or, more generally, the equipment is cleaned out) and the cycle begins anew. Figure 2 displays data collected over a 6-month period from a filter at the Los Alamos Plutonium Facility and shows the temporal continuity described above. Figure 3 summarizes the results of three filters from the uranium dust generation experiments conducted in a glove box. A complete listing of the data can be found in Ref. 3.

In all cases, the holdup accumulation on the filters is well fit by the model

$$h(t) = \alpha t + \beta t^2 \quad ,$$

where h(t) is the amount of holdup on the filter when the throughput is t kg, and α and β are constants. Curves of this form are superimposed on Figs. 2 and 3. The latter figure clearly shows the dependence of the constants α and β on the

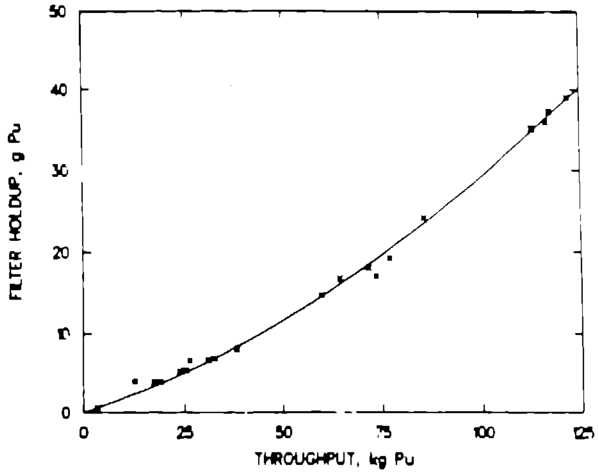

Fig. 2.
Holdup data from a filter at a plutonium processing facility.

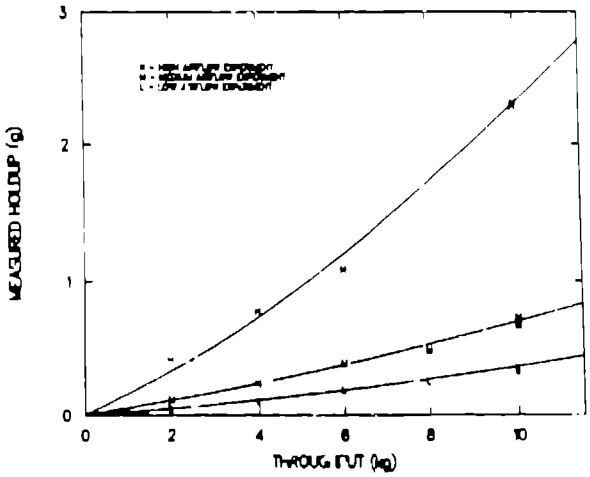

Fig. 3.
Holdup data from air filters used in dust generation experiments.

specific operating conditions involved and demonstrates that a model developed for one set of conditions may not apply under another.

Central to good predictability in these experiments are two factors: the high quality of measurement data and the stable operation of the process. The quality of data is important because large measurement errors can easily obscure the nature of material deposition and make difficult the extraction of a model. If measurements are obtained infrequently, problems are compounded. The second important factor concerns process operation. With respect to Fig. 3, it is not difficult to imagine the results of a hypothetical experiment, the first half of which would be conducted at low airflow and the second half at high airflow. More generally, if the airflow changed often, the increase in holdup would not be nearly as smooth as for the curves of Fig. 3.

These experiments indicate that holdup can be described very well through the use of models. Granted, the controlled experiments represent "best-case" situations and that conditions at facilities are not so idealized. Nonetheless, if adequate importance is attached to estimation of holdup in a particular piece of equipment, measurements of reasonable quality can usually be obtained. When process operation is sufficiently stable, models are quite useful.

Holdup on the filters here is well estimated, even at times when no data are obtained, such as at t = 9 kg in Fig. 3. Moreover, holdup behavior can be accurately predicted for a limited time into the future. When predicting future holdup, there are two important considerations to keep in mind. The first is that it is implicitly assumed that the nature of process operation will remain (nominally) the same as that for which the model applies. The second consideration concerns the nature of the standard deviation of predicted values. As would be expected, the further into the future a prediction is made, the less accurate it is likely to be. Maintaining good estimation requires that measurements be obtained periodically and used to update the fitted model. The frequency of data collection depends on the desired accuracy of estimation.

The procedure for model updating is relatively simple. When a new measurement m(t) is obtained at throughput t, it is compared to its prediction $\hat{h}(t)$ from the model, which is based on previous data. The difference $m(t) - \hat{h}(t)$ should fall within a prescribed range--say, plus or minus three standard deviations of the difference. If so, m(t) is added to the previous data and parameters in the model are re-estimated using all available information. On the other hand, if the difference is too large, this is an indication that the model may have broken down or, perhaps, the measurement is an outlier. In either case, further investigation is suggested.

C. Modeling With Respect to Time: Steady State

Consider the measurement history for the calciner of the ADU experiment at Los Alamos (Fig. 4). Holdup here does not follow the life cycle behavior exhibited for the filters. Instead, beginning from a clean state, a brief initial increase in holdup is followed by long-term fluctuation about steady-state conditions. Process variability plays a major role in estimation: other information concerning the measured values indicates that observed differences in measurements during the steady-state period are not solely the consequence of measurement errors but that the actual amount of material is also changing.

Modeling of steady-state processes is not difficult and typically involves Kalman filtering. This methodology, developed in the early 1960s, has been applied to a wide variety of engineering problems. Applications in safeguards, however, are comparatively few, and it has been suggested[6] that the ostensibly esoteric qualities of Kalman filtering have precluded acceptance by safeguards audiences. If true, this state of affairs need not continue. A major benefit of the Kalman filter is its ability to incorporate process variability; i.e., variability in the actual amount of holdup over time. The measurement history from a poorly measured but stable process might strongly resemble the history from a well measured but unstable process. Thus, holdup estimation crucially depends on the relative magnitudes of measurement errors and normal process variability.

The basis for the Kalman filter lies in the measurement and state equations. For the i^{th} measured value, $m(t_i)$, obtained when the throughput is t_i kg, the measurement equation is

$$m(t_i) = h(t_i) + e(t_i) \quad ,$$

where $h(t_i)$ denotes the actual holdup and $e(t_i)$ is the measurement error. Most models presume $e(t_i)$ is normally distributed with mean zero and standard deviation σ_m. Generally, σ_m can be estimated from measurement control information.

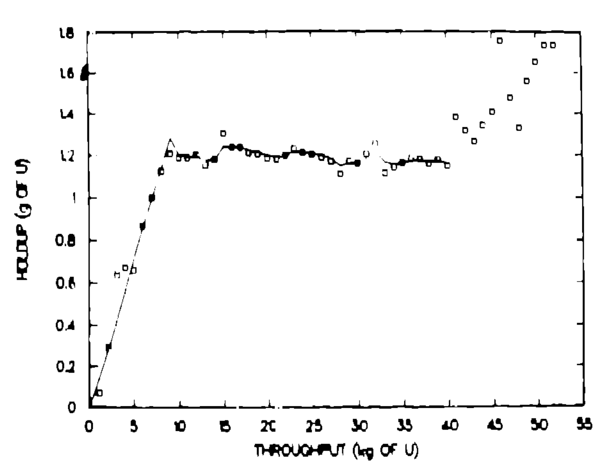

Fig. 4.
Holdup data and model for the calciner.

The state equation is

$$h(t_i) = h(t_{i-1}) + \epsilon(t_i) \quad ,$$

and reflects the steady-state character of the process. The difference $h(t_i) - h(t_{i-1}) = \epsilon(t_i)$ in actual holdup between throughputs t_{i-1} and t_i is assumed to act as a random variable with mean zero and variance $(t_i - t_{i-1})\sigma_p^2$. For the calciner, measurements are obtained with each kilogram of throughput, so that $t_i - t_{i-1}$ is always one. Had measurements been obtained that were unequally spaced, some of the $\epsilon(t_i)$ would have been more variable than others. A simple interpretation is that the change in actual holdup over the interval (t_{i-1}, t_i) is likely to be small if t_{i-1} and t_i are close together, but is likely to be larger otherwise. That $var[\epsilon(t_i)]$ is proportional to the width of the interval (t_{i-1}, t_i) evolves from viewing this interval as a union of smaller, independent subintervals.

The steady-state model outlined here is analogous to the ARIMA(0,1,1) structure of conventional time series analysis. Also, process variability can be incorporated into models where holdup is increasing. More detailed discussions of Kalman filtering can be found in the literature.[7]

Measured values $\{m(t_i)\}$ and estimates of measurement variability σ_m and process variability σ_p are input to the Kalman filter, which produces estimated values of holdup $\{\hat{h}(t_i)\}$. For the calciner, these estimates are connected by line segments in the steady-state portion of Fig. 4. It can also be noticed in Fig. 4 that following the steady-state portion of the data, a marked increase in holdup began after throughput t_{40} = 40 kg. This increase was caused by a change in experimental conditions: the calcining temperature, previously 800°C, was raised to 900°C at that time. The resulting impact on holdup is a vivid indication of how the nature of material deposition can be very dependent on operating conditions.

D. Modeling With Respect To Space

For large pieces of equipment, such as a pulse column or a precipitator it is not possible to accurately estimate holdup at a particular time based on a single nondestructive measurement. The accumulation of holdup can be nonuniform across space; e.g., different sections along the length of a precipitator column can contain different concentrations of material. It is necessary to acquire measurements from different locations to estimate the holdup profile.

As an example, consider the precipitator used in the ADU experiment. At each of 17 locations along the column, concentration measurements (grams of holdup per unit length) were obtained. One such set of data is displayed in Fig. 5, plotted for convenience in log scale.

It is clear that holdup is not uniformly distributed on the interior of the precipitator. Large accumulations in the upper portions of the

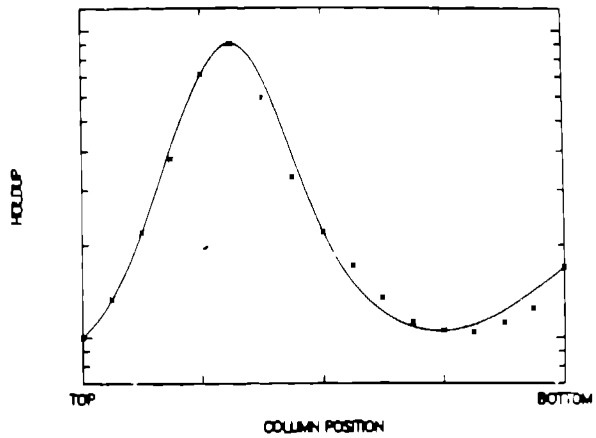

Fig. 5.
Holdup profile of ADU from a precipitator column.

column are caused by violent chemical reactions that lead to phase changes when the NH_4OH contacts the uranyl nitrate solution. Some of the ADU formed at this interface is splashed onto the interior surface above the liquid level. At the bottom of the column, the process of draining the ADU leads to the transport of material there and thus slightly increased residual holdup.

Once an estimated profile is obtained, mathematical integration of that profile provides the estimate of holdup. The same approach can be easily extended to cover material deposited over large two-dimensional areas: fitted contours are developed and then integrated. This type of modeling is analogous to response-surface methodology and is discussed in many statistical texts.

Also, holdup can be modeled with respect to both space and time. This requires estimation of a time-varying profile. Detailed illustration of such modeling and examples are given in Ref. 3. Certain aspects of multivariate time series analysis may be applied to such problems.

V. CONCLUSIONS

The major findings of this investigation are the following:

(1) Measurement of the residual holdup of SNM at large processing facilities is a difficult problem and will remain so because of the inherent limitations of plant layout and NDA techniques.

(2) There are several approaches to improving the quality of measurements involving better instrumentation, better calibration standards, and the application of carefully chosen secondary measurement techniques.

(3) Statistical estimation models can play an important role in materials accounting. Detailed knowledge of process operations, variability of process conditions, and quality of measurements impact the value of model-based estimates.

(4) Significant improvements to holdup measurements and data collection for holdup estimation can be achieved if these problems are addressed during the design stages when new equipment is

installed and the necessary features are incorporated to accomplish the measurement goals.

There are considerable difficulties associated with the measurement and the development of reliable estimates of the holdup of SNM in large processing facilities. Materials accumulating on the surfaces of cracks, pores, and zones of poor circulation of process equipment are not easily measured by conventional methods. This examination of the potential value of developing statistical models that are useful for holdup prediction leads us to conclude that there are many instances in which modeling can be beneficial to developing estimates of the residual inventories of SNM. The value of the statistical model is very much dependent on the quality of the holdup data used in the development of such a model. If the operating conditions are subject to frequent changes and/or the measurement errors are very large, it is unrealistic to expect the development of useful estimation models under such conditions. On the other hand, if the process operation is stable and the holdup data gathered are of good quality, the models developed can be very valuable to making present and future estimates of holdup.

The findings of this investigation also revealed that several factors such as the layout of pipes, corrosion of materials of construction, concentrations of solutions, etc., impact holdup of materials in processing facilities, and in many instances the holdup of SNM is not simply a function of the material throughput.

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