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TITLE HOLDUP-RELATED ISSUES IN SAFEGUARDING OF NUCLEAR MATERIALS

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ABSTRACT

Residual inventories of special nuclear materials (SNM) remaining in processing facilities (holdup) are recognized as an insidious problem for both safety and safeguards. This paper identifies some of the issues that are of concern to the safeguards community at-large that are related to holdup of SNM in large-scale process equipment. These issues range from basic technologies of SNM production to changing regulatory requirements to meet the needs of safeguarding nuclear materials. Although there are no magic formulas to resolve these issues, there are several initiatives that could be taken in areas of facility design, plant operation, personnel training, SNM monitoring, and regulatory guidelines to minimize the problems of holdup and thereby improve both safety and safeguards at nuclear material processing plants.

I. INTRODUCTION

In recent years, because of the increasing concerns over theft/diversion of special nuclear materials (SNM) for clandestine applications, regulatory requirements for safeguarding SNM have become very stringent, almost to the point of seriously affecting the main missions of production facilities. An important feature of current regulations in the U.S is the requirement to report inventory differences (IDs) periodically to regulatory agencies, congressional oversight committees, and the public. This requirement, which came

into effect in 1977, did further encourage bulk-handling facilities, where potentials for SNM holdup are generally high, to examine ways to reduce holdup.

Holdup of materials in process equipment is not unique to SNM processing. But uncontrolled accumulation of materials within process equipment is both a safety and safeguards concern at nuclear material processing facilities. Holdup of SNM in process facilities can result from both normal and abnormal operations of the plant. From a detailed knowledge of the process chemistry and behavior of material forms, it is possible to make reasonable predictions about regions of holdup as well as the relative magnitude of holdup in several kinds of process equipment during normal operations. However, it would be extremely difficult to speculate on the magnitude of holdup during abnormal conditions resulting from process upsets and/or improper plant operations.

II. THE ISSUES

The role of hidden inventories, or holdup, as a safeguards problem is now recognized by almost everyone interested in establishing effective safeguards for SNM. As part of the effort to organize the first INMM-sponsored workshop on process holdup of SNM, a literature survey identified over 70 publications in the open literature relevant to this subject.ⁱ Some of the major issues related to holdup that are of concern at present to the safeguards community are identified in the following paragraphs along with some personal thoughts on the issues themselves and possible approaches to addressing them.

1. Impact of Holdup on Plant and Process Designs

The influence of plant design in SNM holdup at bulk-handling facilities has long been recognized. In the mid-1970s, the U.S. Atomic Energy Agency issued three specific guidelines²⁻⁴ describing desirable design features of facilities for minimizing holdup. Process selection and equipment design influence holdup, and in many instances there are alternatives that can be chosen to minimize holdup. This latter approach has not yet received the attention it deserves. An illustrative example of alternative process design is the use of microwave heating for the direct conversion of uranyl nitrate to uranium oxide. This operation is still carried out in a sequence of steps involving ammonium diuranate precipitation, filtration, drying, calcination to U_3O_8 and subsequent reduction to UO_2 . A significant reduction in holdup can be achieved in this process through the direct denitration and oxidation of uranyl nitrate to UO_2 . Similar process changes can minimize holdup during large-scale processing of all SNM.

A second example is in the use of new materials in facility construction and fabrication of process equipment. Synthetic polymers, such as chlorinated polyvinyl chloride (CPVC), polyvinyl diene fluoride (PVDF) such as "KYNAR," fiberglass reinforced plastics (FRPs), and corrosion-resistant alloys, such as Hastalloy, are now available for large-scale plant construction and equipment fabrication. In addition, pure metals, such as tantalum and surface liners made of high-temperature plastics, are now available at reasonable cost for process facility applications requiring reduced corrosion and surface adhesion.

2. Impact of Holdup on IDs

It is extremely difficult to locate and measure all hidden inventories of SNM in a large plant, and this is the primary reason for large inventory differences (IDs) at some of the bulk handling facilities in the U.S. A rough estimate of the amount of plutonium or uranium required to create a thousandth of an inch (0.025 mm) coating in about 500 miles (800 km) of 1-inch (2.54 cm) diameter pipes (the length of pipe ordinarily found in several large processing facilities) is over 540 kg. This estimate assumes that the deposit has a density of 1 gm/cm³ and that only a third of the deposit is the elemental form of the SNM. In many SNM processing facilities, there are many pieces of equipment with much larger surface areas and those with greater potentials for heavier material deposition.

Large inventory differences have resulted in many kinds of regulatory actions at a number of nuclear material processing facilities in the U.S. They have ranged from a temporary halting of facility operations to complete cessation of operations and decommissioning of facilities. The regulatory agencies have begun to recognize the seriousness of this problem. Beginning in January 1986, the U.S. DOE adopted a new format for presenting its semi-annual report on strategic SNM inventory difference.⁵ The new format clearly demonstrates a recognition of the contributions of process holdup to inventory differences. Of the nine recognized categories of IDs, two of them, "process holdup difference" and "equipment holdup difference," account for a major fraction of total IDs at several bulk-handling facilities.

3. Impact of Holdup on Plant Operations

In addition to large IDs, unidentified holdup of SNM in process equipment is a serious safety problem due to its potential for criticality events. A variety of factors contribute to this situation, including the details of the process, equipment design, personnel training, process upsets, and the facility management philosophy. It is possible to identify and address these issues and take corrective measures to minimize the impact on both safety and safeguards.

4. Periodic Cleanout of Equipment for Materials Accounting

Although periodic termination of process operations to cleanout equipment for materials accounting is a desirable goal for good materials accounting, this practice is counter-productive to the primary missions of such facilities, namely the production of nuclear materials economically and efficiently. Therefore, such practices are frowned upon by managers of process facilities. As a result, most holdup measurements done to-date have been in response to large IDs or criticality safety concerns. Prudent management of facilities processing large amounts of highly enriched uranium and separated plutonium should include scheduled cleanout operations.

5. Advantages and Limitations of Nondestructive Assay (NDA) Techniques for Holdup Estimation

NDA techniques for measuring plutonium and uranium, using passive gamma and neutron measurements, have continually improved over the past two decades. There are, however, fundamental limitations to these NDA techniques that will continue to affect holdup measurements. The crowded environments of process

facilities, combined with non-stoichiometry and nonuniformity of holdup residuals and the inadequacies of calibration standards and measurement equipment designs, will continue to offer challenges to NDA measurements of holdup.

6. Development of NDA Instruments and Standards for Holdup Measurement

Developing suitable standards for NDA of radioactive materials is always a challenge. However, in the case of holdup measurements, the problem becomes extremely complex due to the nonhomogeneity of the sample to be assayed, its unknown distribution pattern, varying chemical composition, the complex geometry of the equipment in which the materials reside, the attenuation of radiations by the equipment and the matrix, and the high background radiation levels in processing areas. Ideally, it is desirable to have calibration sources closely simulating the actual holdup deposits to be assayed. The present practices are to use point sources, line sources, or uniform flat sources as calibration standards and extrapolate the results to other complex geometries. Although this approach is adequate in many instances and is a desirable compromise at others, there is much that can be done to improve calibration standards for holdup measurements and thus minimize the uncertainties of such estimates.^{6,7}

7. Regulatory Reforms to Accommodate Holdup Estimates to Reduce IDs

Although safeguarding of SNM was always considered important from the early days of nuclear technology development, regulatory guidelines to achieve this objective have been in a state of evolution. There is a growing recognition that a significant part of the IDs at the bulk handling facilities is due to unmeasured inventories and/or holdup. In addition to regulatory pressures

to reduce IDs, there are several safety and safeguards issues related to cumulative effects of SNM holdup. Because of the uniqueness of nuclear criticality safety, many holdup-related safety issues are addressed during plant and process designs. However, in the past, safeguards issues seldom influenced facility design, and they are often difficult to resolve later.

8. Pragmatic Alternatives to Plant-Wide Holdup Measurements

A prevailing view is that facility-wide holdup estimates would have very large measurement and sampling errors; thus, adjusting an ID to reflect an estimated change in holdup would create a new quantity with uncertainty so large as to render the quantity meaningless. An objective re-examination of this view as well as current regulations and the development of pragmatic approaches to include sound estimates of holdup in calculating IDs would be a step in the right direction.

Regulatory reforms encouraging the use of modern tools, such as sampling and modeling, to maximize the use of resources and develop technically sound estimates of holdup without interrupting production schedules would encourage facility operators to invest resources to address this problem. Although indirect methods of holdup estimation⁸ using tracer techniques and mathematical modeling have been demonstrated to be viable and less intrusive, there have not yet been any plant-wide applications of these approaches.

9. Costs and Benefits

Current regulatory guidelines do not offer sufficient incentives to facility operators to invest resources to estimate holdup. Because various nuclear

fuel cycle facilities contain materials of various attractiveness, the safeguards and resource requirements do differ considerably across the nuclear materials production complex. Large-scale resource investments to minimize holdup and improve safeguards at fuel cycle facilities handling materials of low attractiveness would not be prudent. At the same time, it would be a good investment to spend adequate resources to minimize holdup and improve safeguards by addressing all the issues discussed in this paper, including an occasional shut-down for a complete cleanout inventory.

10. Research Efforts to Address Specific Holdup Problems

In the nuclear fuel cycle, there are very few areas where holdup of SNM is not a problem. However, the issues are of great concern when the materials are in chemically pure and isotopically enriched form due to attractiveness of materials as well as their potential for causing criticality accidents. Presently, there are several on-going projects in the U.S. to refurbish and renovate aging production facilities as well as to build new ones. These projects offer unique opportunities to incorporate state of the art technologies that would minimize holdup problems and improve overall efficiency of materials production. In areas of holdup measurement, there is a crying need to develop specially designed monitoring equipment and calibration standards to meet the special needs of holdup measurement. Indirect measurement capabilities, innovative calibration techniques, portable assay equipment, and proper personnel training can go a long way in alleviating holdup-related problems at SNM production facilities.

III. SUMMARY

There is a growing recognition that holdup of SNM in processing facilities is deleterious not only to safety but to the safeguarding of such materials. Both facility operators and regulatory agencies are beginning to address the complex issues that have been kept in abeyance for a long time. Presently, there are new opportunities and challenges to develop and apply new processes, equipment designs and materials, innovative plant layouts, and specially designed radiation measurement instruments and calibration standards. Open discussion of issues identified here is an essential step in addressing holdup-related issues rationally and to develop satisfactory solutions to a problem that has plagued the nuclear process industry for the past four decades.

IV. REFERENCES

1. K. K. S. Pillay, "A Survey of Open Literature Publications on Process Holdup of Special Nuclear Materials," LA-UR-87-4185, in Summary of Papers presented at the Technical Workshop on Process Holdup of Special Nuclear Materials, Rockville, MD (March 1988).
2. Regulatory Guide 5.8, "Design Considerations for Minimizing Residual Holdup of Special Nuclear Material in Drying and Fluidized Bed Operations," US Atomic Energy Commission (May 1974).

3. Regulatory Guide 5.25, "Design Considerations for Minimizing Residual Hold-up of Special Nuclear Material in Equipment for Wet Processing Operations," US Atomic Energy Commission (June 1974).
4. Regulatory Guide 5.42, "Design Considerations for Minimizing Residual Hold-up of Special Nuclear Material in Equipment for Dry Process Operations." US Atomic Energy Commission (January 1975).
5. "Semiannual Report on Strategic Special Nuclear Material Inventory Difference," DOE/DP-001/17 (January 1986).
6. K.K.S. Pillay, "Tracer Techniques in Estimating Nuclear Materials Holdup," J. Radioanal. Nucl. Chem (1988).
7. S. F. Marsh, "Nondestructive Assay of Plutonium Residue in Horizontal Storage Tanks," in "Analytical Chemistry Instrumentation," Ed. W. R. Laing, Lewis Publishers, New York, NY, 91 (1986)
8. K.K.S. Pillay, R. R. Picard, and R. S. Marshall, "Estimation Methods for Process Holdup of Special Nuclear Materials," LA-10038, Los Alamos National Laboratory, Los Alamos, NM (June 1984).