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SAFEGUARDS IMPLEMENTATION IN THE NUCLEAR FUEL CYCLE

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1. INTRODUCTION

Recent nuclear power projections of various nations[1] conservatively indicate that some half-a-million megawatts of nuclear electric generating capacity will be commissioned around the world within the next ten years. To fuel this nuclear capacity will require the production of more than 75,000 tons of natural uranium annually, some 55 million SWU to enrich it, and more than 12,000 tons of fuel will have to go through fabrication and some form of spent fuel disposition (e.g., storage, reprocessing, or some combination of these).

The challenge of how to strengthen and foster the worldwide growth of nuclear power while at the same time decreasing the accompanying risks of nuclear diversion, proliferation, etc., is being addressed by the International Fuel Cycle Evaluation (INFCE) Program and related fuel cycle studies presently underway in a number of countries. Whatever the results of INFCE, it seems clear that world nuclear power demands will, in the near future, require high-throughput process facilities to support any of several alternative fuel cycles (or "mix" of fuel cycles) that are selected for implementation in various countries. And, since very large quantities of strategic nuclear materials will be involved, nuclear safeguards considerations are becoming a major factor in the selection of process and facility design/construction alternatives. Today's trend toward tightening regulations and increasingly stringent safeguards -- in both the overall international (IAEA) system, and the various component State (national) systems -- further underscores the necessity for nuclear safeguards criteria to be fully incorporated at an early stage in the design of future fuel cycle facilities or centers - be they national, regional, and/or international.

In the nuclear energy area particularly, the vital importance of international cooperation, exchange, and understanding -- as exemplified by this biannual series of Pacific Basin Fuel Cycle Conferences - has been stressed by many leaders throughout the world nuclear community[1,2,3]. Specifically in the field of nuclear safeguards it has been repeatedly emphasized[4,5] that today's mounting demands for high-technology safeguards systems (demands in terms of both financial and human resources) will, of necessity, require closer international cooperation and increased technical exchange in the design, test, and evaluation of advanced safeguards technology and control systems.

II. IAEA INTERNATIONAL SAFEGUARDS AND NATIONAL SAFEGUARDS SYSTEMS

At the 21st Session of the General Conference of the International Atomic Energy Agency, the Director General of the Agency, Dr. Sigvard Eklund[9], in his opening remarks on safeguards and nonproliferation, noted that the IAEA is itself a product of man's awareness of the dichotomous nature of nuclear energy -- i.e., it can contribute significantly to the fulfillment of vital economic and

social goals through almost unbounded production of energy, or alternatively, it can also provide the source of unprecedented destruction capability. Eklund further observed that "safeguards (in the sense of international measures to detect and thereby deter the diversion and misuse of nuclear materials) remain the central element of any combination of measures taken against nuclear proliferation; their existence has been shown to be a primary condition for international commerce and cooperation in the nuclear field. . . .International interest in the potential effectiveness of safeguards continues to increase, . . .and intensive development work will be essential to make safeguards both more credible and more cost effective.... I need hardly mention that the support we are receiving from member states is absolutely essential for these programmes."

Effective national safeguards systems are indeed essential components of effective international (IAEA) safeguards, which are in turn essential to the widespread growth and acceptability of nuclear power, and the concomitant worldwide expansion of nuclear trade. At the IAEA Salzburg Conference on Nuclear Power and its Fuel Cycle[1], leading IAEA officials stressed that notwithstanding the requirements for improved national and multi-national systems of nuclear materials accountancy and control, the sine qua non of effective international safeguards is independent verification by the IAEA of compliance with the provisions of safeguards agreements concluded pursuant to the NPT and to the Statute of the IAEA. It was further emphasized that this independent verification is the basis of the IAEA safeguards system and this responsibility cannot be transferred to any other authorities.

The importance of continued technical developments and full-fuel-cycle safeguards implementation both in member states and internationally by the IAEA has also been stressed by Eklund (among many others) at Salzburg[1] and elsewhere[9,10], along with specific reference to the role of integrated material accountancy systems, "real time" material control systems, containment and surveillance methods, and modeling techniques for evaluating the effectiveness of modern safeguards systems. In various member states of the IAEA a growing number of safeguards development test and evaluation programs are underway, and more are planned. As one component part of this overall effort to enhance both national and international safeguards, we shall review here some representative major programmatic activities in the United States in the areas of safeguards system design, technology development, test, evaluation and in-plant implementation.

III. REFERENCE FACILITY SAFEGUARDS SYSTEM DESIGN

In the United States, as a part of the U. S. Department of Energy, Safeguards and Security R&D program[11,12] conceptual designs of integrated safeguards and materials management systems have been developed and evaluated for the major components of the back end of the LWR fuel cycle[13,14]. These designs and safeguards performance criteria are being used as the reference designs in the evaluation of similar facilities in alternative fuel cycles, presently under study in INFCE and related programs. These designs incorporate state-of-the-art materials accounting systems that can be closely integrated[15] with advanced physical protection systems[16-19] to provide overall facility safeguards effectiveness and minimum interference with plant operation, efficiency, and throughput.

To date, conceptual designs of materials management and accountability systems (MMAS) have been completed for a LWR fuel fabrication plant[20], a nitrate-to-oxide conversion plant[21], a large scale chemical separation facility[22] and a nuclear criticality facility[23]. Fuel storage and waste management facilities are also under study. Each material accounting system design is based on a specific reference facility so that realistic and quantitative

conclusions can be reached. The Westinghouse, Anderson design was selected as the reference facility for mixed-oxide fuel fabrication[24]. The conversion facility is based on a reference design by the Savannah River Laboratory and Savannah River Plant[25]. The Barnwell Nuclear Fuel Processing Plant at Barnwell, South Carolina[26] was selected as the reference chemical separations facility.

IV. CONVENTIONAL MATERIALS ACCOUNTING

In conventional safeguards practice, the accountability of nuclear materials within a facility and the detection of unauthorized removals have relied, almost exclusively, on discrete-item counting and material-balance accounting following periodic shutdown, cleanout, and physical inventory. The classical material balance is usually drawn around the entire facility or a major portion of the process, and is formed by adding all measured receipts to the initial measured inventory and subtracting all measured removals from the final measured inventory. During routine production, material control is vested largely in administrative and process controls, augmented by secure storage for discrete items, sealed containers, etc.

Although this basic method of material-balance accounting is essential to safeguards control of nuclear materials, the conventional procedures used in the past have inherent limitations in sensitivity and timeliness. Sensitivity is limited by measurement uncertainties that might obscure the diversion of relatively large quantities of SNM in a large-throughput plant. Timeliness is limited by the practical difficulties, the expense, and hence the infrequency, of process shutdown, cleanout, and physical inventory; i.e., a loss of material could remain undiscovered until the next physical inventory is taken.

V. DYNAMIC MATERIAL ACCOUNTING/CONTROL

Recently developed nondestructive assay (NDA) technology, state-of-the-art conventional measurement methods and special in-plant sensors, combined with supportive computer and data-base management technology have provided the necessary technical basis for much more effective alternative methods of safeguarding nuclear facilities. It has been demonstrated for example, that considerably greater sensitivity and timeliness in SNM control can be achieved by subdividing a nuclear facility into discrete accounting envelopes, called unit processes, around which individual balances can be drawn[27,28]. A unit process can be one or more chemical or physical processes, and is chosen on the basis of process logic, residence time of material within the unit process, and the ability to perform quantitative measurements and draw a material balance. Thus, by subdividing a facility into unit processes and measuring all material flows across unit process boundaries, the location and movement of SNM throughout the plant can be localized both in space and time. Material balances drawn around such unit processes are called "dynamic" material balances to distinguish them from conventional balances drawn after a shutdown, cleanout, and physical inventory. As is the case with any material balance based on physical measurements, balances cannot be closed completely (i.e., MUF = 0 precisely); this partly due, of course, to ever-present measurement uncertainties, but also because some in-process holdup and minor sidestreams are customarily (and generally for good reason) measured less frequently than major materials transfers[5,28,9]. Such perturbations are normally handled quite adequately by using plant operational experience or "historical" data to interpret trends in holdup, minor sidestreams, etc., with these data being updated when the appropriate holdup and sidestream measurements are made. In some processes, the added control obtained by measuring small sidestreams of material may be negligible, and may not justify the difficulty and expense of making the measurements. Such judgements must, of course, be made on an individual process basis, taking into account "graded safeguards" considerations

of the strategic value and safeguards vulnerability of the material, which will depend on its location and form within the process and within the fuel cycle[13]. Graded safeguards considerations are clearly important in the selection of unit processes and associated key measurement points.

Implementation of dynamic or "near real time" materials measurement and control requires the rapid, quantitative measurement of nuclear materials locally (e.g., in-line or at-line) at each unit process. Modern nondestructive assay techniques are quite well suited to rapid, direct in-line measurement, and NDA instruments are being developed, adapted and applied to process measurement requirements in several different ways: (1) as the primary measurement technique at a unit-process boundary, (2) as part of a complementary set consisting of timely, or even "continuous", NDA measurements that may be updated by periodic analytical chemistry assay, (e.g., in conjunction with shutdown and cleanout, as appropriate) and (3) to assay or verify the contents of discrete items such as sealed containers, fabricated pieces, finished components, etc. (where the nondestructive feature of the assay is particularly advantageous).

As may be inferred from the foregoing, current trends in nuclear safeguards technology[8] place increasing emphasis on timely measurement and analysis of materials accounting data of constantly improved quality. The availability of more and better input data underscores the need for an organized framework of techniques to ensure efficient and complete extraction of information concerning possible diversion of SNM. The discipline of decision analysis[30] which combines techniques from estimation theory, decision theory, and systems analysis, provides such a framework, and is well suited for statistical treatment of the imperfect material-balance data that become available sequentially in time. The goals of decision analysis are (1) detection of the event(s) that SNM has been diverted, (2) estimation of the amount(s) diverted, and (3) determination of the significance of the estimates. Augmented by pattern-recognition techniques such as the V-mask test for Cusum plots and the alarm-sequence chart, decision analysis can be used to reduce errors caused by subjective data evaluation and to condense large collections of data to a smaller set of more descriptive statistics. The use of these powerful, formalized techniques make the decision process more timely and efficient as well as more consistent and objective[31].

The availability of advanced NDA measurement technology, together with in-line process instrumentation, automatic item identification and verification equipment, as well as modern computerized data analysis and data base management technology, has led to a whole new generation of dynamic materials accounting/ control systems that are currently in various stages of development. The degree of complexity of the different systems ranges from simple computerized accounting systems (e.g., for discrete item control) to complete deployment of each of the advanced technologies noted above. Some of the more sophisticated materials control and accounting systems being developed are summarized and documented in Table I.

In the United States, the DYMAC* program[27,47-50] at the Los Alamos Plutonium Processing Facility represents one of the most extensive R&D efforts to integrate advanced NDA technology with automated data-processing methods and to fully evaluate practical in-plant operation of dynamic material accounting and control on a detailed unit-process basis. A number of NDA measurement systems are being developed, or adapted from existing designs, commercially available equipment, etc., for in-line DYMAC applications in the new plutonium processing facility at TA-55, LASL. These include:

*DYMAC is the acronym for Dynamic Materials Control, or equivalently, for Dynamic Materials Accounting.

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TABLE I.

SOME DYNAMIC MATERIALS ACCOUNTING SYSTEMS

Facility, Location, Function	System Name	System Functions and Comments	Reference
General Electric Wilmington, North Carolina; UF$_6$ conversion to fuel-bundle assembly	MICS	Material Inventory Control System. Diversion detection, information quality, loss localization, system management and control. NDA used.	32,33
General Electric Vallecitos, California; Pu fuel-development laboratory for LMFBR	GERTA	Material distribution, diversion detection. Primarily automated record-keeping; no NDA at this time.	34
Mound Laboratory Miamisburg, Ohio; MOX fuel fabrication	CUA	Controlled Unit Accounting. Conceptual system for accounting, diversion detection. No NDA used.	35,36
Combustion Engineering Windsor, Connecticut; Nuclear fuel manufacturing	FACS	Fuel Accounting and Control System. Timely and accurate reporting on SNM status and flow; no NDA as yet.	37
AECL Chalk River, Canada; Fuel materials development and fabrication	INMACS	Integrated Nuclear Material Accounting and Control System. On-line material accounting, data base mgmt.; no NDA as yet.	38
Y-12 Plant Oak Ridge, Tennessee; Enriched uranium processing facility	DYMCAS	Accountability, diversion detection, physical inventory, NDA verification. Incorporates on-line or keyboard verification of weights.	39,40

TABLE I. (cont.)

Facility, Location, Function	System Name	System Functions and Comments	Reference
Rocky Flats Plant Golden, Colorado; Pu processing facility	NMC COMSAC	Accountability, criticality control, NDA calibration, NDA measurements.	41,42
Karlsruhe Research Center Karlsruhe, F.R.G.; Research, processing, handling, storage facilities	---	Generalized SNM accounting and data handling system for variety of SNM processing, handling functions.	43
ARHCO Richland, Washington; Storage and processing facility	---	Accountability, process monitoring, laboratory bookkeeping, monitoring and control of storage locations.	44
PNC Tokai-mura, Japan; MOX fuel fabrication	PINC	Plutonium Inventory Control system using on-line NDA sensors and computerized inventory, process control.	45
AGNS Barnwell, South Carolina; Fuel reprocessing plant	AGMAC	Laboratory data system and materials accounting and control system. Some process monitoring.	46
LASL Los Alamos, New Mexico; Pu processing	DYMAC	Accountability, in-plant NDA instrumentation; computerized near-real-time inventory control; data-base management, unit process SNM localization.	47-50

(1) a plutonium solution assay system (PUSAS) for measuring Pu concentrations over the range 0.1-20 g/L;

(2) a specialized thermal neutron coincidence counter (TNC) for measuring residues from various recovery processes;

(3) a fast neutron coincidence system for NDA of heterogeneous materials having high (α,n) backgrounds;

(4) high resolution gamma spectrometry for isotopic verification;

(5) product verification stations; and

(6) a variety of digital readout weighing devices.

Design philosophy on NDA instruments used for in-line plutonium assay has been to keep delicate parts of the instrument (e.g., detector and electronics) outside gloveboxes whenever possible. In designing or adapting instruments for glovebox use, careful consideration was given to the frequently cumbersome and awkward nature of having to work through gloves. By focusing attention on the specific functional use of the instrument and working closely with process operators during instrument development, it was possible to maximize operational convenience while minimizing required operator time for making the assay. DYMAC in-plant experience to date[49] has demonstrated that process operators are receptive to entering material transactions at interactive computer terminals in the process area, provided the time and number of entries necessary to complete a transaction are reasonable. In order to streamline the transactions for process areas and specialize them to reduce the amount of information the operator must enter, a flexible "packet software" approach was developed that enables appropriate changes to be readily programmed for any specialized process transaction.

Figure 1 shows the location of DYMAC instruments and terminals in the plutonium recycle wing of the Los Alamos plutonium facility (TA-55). An integral part of the DYMAC system is a rigorous standards and measurement control program[49,50] that assures the accuracy of the assay data. The program provides quantitative limits-of-error information and ensures that the individual instruments function properly by periodically checking their precision and calibration accuracy.

The basic structure of the DYMAC information handling system is shown in Figure 2. The NDA instruments and interactive terminals in each unit process area transmit the status of SNM in the various stages of production to a central computer system which acts as the central data manager for DYMAC. In the computer (cf. Fig. 2), the data base management subsystem accepts and verifies incoming data, updates inventory records immediately, and organizes the data into files for efficient retrieval of specific information. The real time materials accountability (control) subsystem draws on the data base for continuous status monitoring of SNM within the facility. A measurement control program periodically checks instrument performance; control parameters are calculated from information in the data base and are compared with pre-determined alarm levels. If alarm levels are exceeded, the system alerts the nuclear materials officer -- or "safeguards coordination unit" (cf. Ref. 20, Figure III-1). As a focal point for safeguards decisions, the safeguards coordination unit interacts with management and process-control coordination to continually assess the safeguards status of the plant and to advise plant management of appropriate response options and recommended actions. Needless to say, safeguards condition assessment and associated decision analysis procedures[31] must be carefully balanced to avoid unnecessary false alarms, while

maintaining a high probability of effective response to an actual safeguards violation.

A key consideration in the design of an acceptable dynamic materials control system is the scrupulous avoidance of any significant intrusion into process operations and overall plant production. The workability and effectiveness of any system can be convincingly demonstrated only through extensive inplant operation and evaluation, as is currently in progress for a number of the systems in Table I.

VI. PROCESS SIMULATION AND SAFEGUARDS EFFECTIVENESS EVALUATION

Somewhat unique in the nuclear fuel cycle, the conversion process (i.e., converting plutonium nitrate solution to plutonium oxide powder) presents a particularly challenging safeguards problem[21]. The conventional conversion facility handles plutonium in large quantities as a concentrated, relatively pure material with generally low radiation levels, and is therefore extremely attractive as a target for diversion. Fortunately, these same attributes also make a conversion facility amenable to stringent safeguards, i.e. such features as well characterized, relatively pure materials and low radiation levels with resulting greater accessibility to the process, all facilitate on-line measurement and the full implementation of dynamic materials accounting and control.

Modeling and simulation techniques[5i] have proved extremely valuable in the design, evaluation, and comparison of the relative effectiveness of alternative processes, measurements systems and materials accounting strategies. These techniques permit the prediction of the dynamics of SNM flow under a wide range of operating parameters, and the rapid accumulation of data for relatively long operating periods. For each facility, this approach requires: (1). a detailed dynamic model of the process; (2). simulation of the model process on a digital computer: (3). a dynamic model of each measurement system; (4). simulation of accountability measurements on SNM flows and in-process inventories generated by the model process; and, (5). evaluation of the simulated measurement data from each accounting strategy.

As a specific example of the performance of dynamic materials accounting, in a modern highthroughput conversion facility, we cite recent simulation studies[21] on a reference conversion process based on plutonium (valence III) oxalate precipitation and calcination. In this conversion process, the key measurement points were located at the receipt tank, the output of the precipitator and at the product loadout area. At the receipt tank the solution volume and concentration are measured, the concentration being measured by an absorption edge densitometer (to be described later). The product canisters containing plutonium oxide powder are measured by a neutron well counter or calorimeter.

The estimated plutonium detection sensitivity levels (for a single-unit-process accounting strategy) are presented in Table II. Diversion sensitivity is given for periods of one material balance (one batch), one day (approximately 20 batches), one week (approximately 125 batches), and one month (approximately 530 batches). The results in Table II may be compared with current U. S. regulations[52] which require that conventional periodic material balancing in conversion plants be performed every two months with a material balance uncertainty (2 σ) of less than 0.5% of the facility throughput. This limit of error corresponds to 33 kg of plutonium for the reference conversion process, which has a design throughput of 6600 kg of plutonium over a two month period. A recent estimate[53] of the 2 σ limit of error that should be achievable by periodic, two month material balancing in conversion plants is 0.38%, which corresponds to approximately 25 kg of plutonium for the reference process.

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TABLE II.

DIVERSION SENSITIVITY[a] FOR THE CONVERSION PROCESS

Detection Time	Average Diversion per Batch (kg Pu)	Total at Time of Detection (kg Pu)
1 batch (1.35 h)	0.13	0.13
1 day	0.03	0.63
1 week	0.01	1.24
1 month	0.005	2.65

---

[a] For a single-unit-process accounting strategy as described in Ref. 21.

These and other process simulation studies[11,20,22] have clearly demonstrated that dynamic material accounting can offer dramatic improvement in terms of timeliness, spatial specificity and sensitivity, when compared to conventional materials accounting procedures. In the final analysis, of course, the effectiveness of any system must be fully demonstrated and proven out through extensive in-plant operation and evaluation by the process, quality control and materials management people who must use (i.e., "live with") the system on a day-to-day basis.

From the safeguards and nonproliferation standpoint, one of the more prominent alternate fuel cyles currently under consideration involves the coprocessing of both uranium and plutonium (in a ratio of roughly 6-10:1) in a fuel reprocessing facility. In general, alternative conversion processes that yield a product usable only as reactor fuel are clearly of potential interest from the safeguards standpoint. One conversion process, Coprecal[54], has been developed specifically for production of mixed uranium-plutonium oxides for fast breeder reactor fuels, and should be ideally suited for coprocessing applications. The Coprecal process converts a coprocessed U/Pu nitrate solution to a mixed oxide powder through Coprecipitation followed by Cocalcination. The integrated safeguards system structure is similar to the three studies referred to above[13,20,22]. The modeling and simulation approach used previously has been applied to the design and evaluation of a material measurement and accounting system for the Coprecal facility[55]. Dynamic material balances can be drawn approximately every two hours about unit processes as well as the whole process. It is shown that dynamic materials accounting as compared to conventional materials accounting, can detect diversion in days or hours instead of months, can localize diversion to a single unit process accounting area instead of the whole process, and can markedly improve diversion sensitivity.

VII. MEASUREMENT TECHNOLOGY R&D - RECENT DEVELOPMENTS AND TRENDS

The implementation of dynamic materials accounting and control relies heavily on modern measurement technology, with particular emphasis on new NDA techniques and in-plant instrumentation. In this section, we review some of the recent developments and trends in SNM measurement technology and NDA instrumentation.

For NDA of plutonium in general, and particularly for solutions, passive gamma-ray assay has proved very useful, primarily for the determination of ^{239}Pu, ^{241}Pu and ^{241}Am isotopic concentrations. When isotopic composition is known, or determined independently, corrections can be applied to yield overall plutonium concentrations to better than 1%[56].

The increasingly popular technique of absorption edge densitometry[57] offers another very versatile method for NDA of solids as well as on-line measurement of actinide concentrations in process streams. The method is based on the difference in transmission of gamma rays with energies just above and just below the K and L_{III} absorption edges, which are uniquely characteristic of the elements uranium, plutonium and thorium. The absorption edge method, being based on the discrete electron binding energies in the electron shell structure of the atom, is thereby element specific, rather than isotope specific as is the case with passive gamma rays emitted from the nucleus. The transmission source used for absorption edge densitometry may be either an x-ray generator[58] or natural radioactive isotope(s)[57]. The x-ray generator has the advantages that: (1). multiple, simultaneous SNM determinations such as plutonium and uranium are possible and (2). the energy displacement from the absorption edge is limited only by the detector resolution. An x-ray generator-based absorption edge densitometry assay station has been developed at Los Alamos for rapid, simultaneous measurement of multiple concentrations of SNM and source materials[59]. The station forms the basis for development, test and evaluation of an

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on-line solution assay system for measuring uranium and plutonium concentrations of 1 to 50 g/liter. Such a system has been proposed for installation and in-line evaluation at the experimental coprocessing test location at the Savannah River Laboratory. The need for incisive measurement technology for different combinations of fissile and fertile fuel materials is underscored by the current interest (e.g., in INFCE) in coprocessing as one of several alternative fuel cycle possibilities for enhancing safeguards in the back end of the fuel cycle.

The assay of multiple SNM by the absorption edge densitometry method can be conveniently performed when the two fissionable components are present in roughly equal amounts (ratios between one and four). For concentration ratios greater than four, the densitometer will spend most of the limited pulse-processing time on the major component (e.g., uranium in a coprocessed stream), leaving the minor component (e.g., coprocessed plutonium) with poor statistics and thus poorly determined. The close proximity of the uranium and plutonium L_{III} edges (17.168 keV and 18.066 keV, respectively) also limits the energy range between the two L_{III} edges, in which the relevant transmissions can be measured.

One approach[59] to improve the measurement statistics is to utilize as much data as possible. A typical x-ray spectrum transmitted through a uranium- and zirconium-bearing solution is shown in Fig. 3(a). For experimental convenience, zirconium can be used to simulate plutonium because the zirconium K edge at 17.998 keV is representative of the plutonium L_{III} edge at 18.066 keV. Selected regions of data have been linearly extrapolated, as shown, to the absorption edges to determine the transmission ratios as shown in Fig. 3(a).

By essentially differentiating the transmission curve (i.e., taking the difference in $\ln(T)$ at each discrete energy step, i, the plot shown in Fig. 3(b) is obtained. Over the narrow energy range of interest, the constant matrix material effects cancel out and the net area under each "peak" in Fig. 3(b) is proportional to the density of the respective SNM, i.e., summing over the N channels of the "peak" in Fig. 3(b):

$$\sum_{i=1}^{N} \ln \frac{T_i}{T_{i-1}} \simeq \Delta\mu_s \, \rho_s \, X$$

where $\Delta\mu_s$ is the discontinuity in SNM mass absorption coefficient across the absorption edge, ρ_s is the density of SNM and X is the thickness of the solution sample.

This approach[59] of using the "peak" area as a measure of the SNM content has the distinct advantage that more data points are utilized in the data analysis, thereby resulting in a more precise determination. For a solution containing 6.2 g Zr/ℓ and 37 g U/ℓ, repeated runs of 1000-s counting time have shown that the uranium content can be measured to one-half per cent and the zirconium content to two per cent.

While absorption-edge densitometry techniques can provide accuracies of 1% or better for solution concentrations above ~5 grams per liter, measurement uncertainties increase to greater than a few per cent for sample concentrations below 2 g/ℓ. To achieve higher precision in the NDA of lower-density solutions, new techniques of energy-dispersive x-ray fluorescence (XRF) are being investigated at Los Alamos[60]. Like absorption edge densitometry, XRF also measures the total elemental concentration. However, by judicious choice of the isotopic source (e.g., ^{109}Cd with a 45-d, 88 keV γ ray) used to induce particular fluorescence x rays characteristic of actinide elements, the need for an x-ray generator can be eliminated, thus reducing the cost and simplifying the design of NDA equipment for solution measurements at lower concentrations.

This technique involves L X-rays typically in the 10-20 keV range and hence requires a method of correcting for sample attenuation effects on both the incoming (inducing source) radiation and the outgoing (induced fluorescence) x-rays. A recent innovation[60] in transmission corrected x-ray fluorescence measurements employs an appropriately selected transmission foil whose induced fluorescence x-rays bracket the characteristic line(s) from the actinide element to be analyzed. Measurements to date indicate that with appropriate combinations of source and transmission foil, assay accuracies and precisions of better than 1% should be routinely achievable on low-concentration solutions (e.g. from ~10 g/ℓ down to ~0.5 g/ℓ).

The two somewhat complementary NDA techniques just described -- i.e., absorption edge densitometry in the concentration range 2 g/ℓ to 50 g/ℓ and transmission corrected x-ray fluorescence .or lower concentrations -- provide a versatile, relatively simple, and accurate means for assaying SNM-bearing solutions found in modern high-throughput process facilities. The importance of possible future applications of these NDA techniques to coprocessed U and Pu solutions, or to alternative fuel cycle mixtures such as U and Th, or U, Pu and Th scarcely needs elaboration.

For highly radioactive solutions, the background suppression already inherent in the energy-selective absorption edge technique may be further enhanced by using a curved crystal spectrometer as an energy filter (with a few keV width, centered around the absorption edge of interest) for a high resolution energy dispersive detector[61]. Further, development test and evaluation will be required to determine the value of this approach to NDA of highly radioactive solutions under plant conditions.

One area that continues to present an in-plant measurement challenge is the determination of in-process holdup. Plant holdup measurements in specific locations are frequently made using passive gamma-ray methods[49,62], and more recently an integral neutron detection method has been used to measure total plutonium holdup in an entire process room[63,64].

Neutron coincidence counting[65] has found wide application in the assay of bulk plutonium product, scrap and waste. The net coincidence count rate is approximately proportional to the weighted mass of ^{240}Pu plus other even plutonium isotopes. If the isotopic composition of the sample is known, or independently determined, e.g. from GeLi spectrometry, then coincidence counting can be used to determine total plutonium content. For coincidence counting of large amounts of PuO_2 or scrap containing light elements having high α,n yields such as boron or fluorine, coincidence counters with short die-away times have been developed to maximize the ratio of real coincidence events to accidental events, and thereby reduce statistical uncertainties. Recent R&D on neutron coincidence counters has been directed toward upgrading assay capability for large plutonium samples. Improvements have been made in the detectors, moderators, and coincidence circuitry to give shorter die-away times and coincidence gates, and decreased electronics deadtime. This has permitted accurate assay of high mass (e.g., ~2 kg) plutonium samples with counting rates on the order of 10^5 counts/second. To accommodate a dynamic range of measurement from less than one gram to greater than 2 kg, a dual range coincidence counter employing removable cadmium sleeves has been developed[66]. A practical limitation on coincidence counting in the high mass range is the accuracy with which required sample self multiplication corrections can be applied[67].

Representative precisions and accuracies exhibited by neutron coincidence counters in the process environment are shown in Table III for the general categories of product, scrap and waste[56].

TABLE III

TYPICAL NEUTRON COINCIDENCE COUNTER UNCERTAINTIES

Material Category	Precision (%)	Accuracy (%)
Feed and product	1	1
Scrap	2-8	2
Waste	10-15	5-10

A [252]Cf "Shuffler" assay system[68] based on neutron interrogation and delayed neutron counting has been evaluated in the laboratory for bulk samples containing uranium and/or plutonium. This unit has be. adapted to the assay of large (55 gal) barrels of hot radioactive waste[69] an. to the assay of uranium feed materials as well as scrap and waste from the reactor fuel (U-Al) fabrication at the Savannah River Plant[70]. The Shuffler can also be used for the measurement of [234]U, [235]U, and plutonium over a wide mass range (1 mg - 2 kg) using thermal-neutron interrogation for the low mass range and fast-neutron interrogation for intermediate and high mass samples. A prototype shuffler system is being designed for test and evaluation at the Idaho Chemical Processing Plant where it will assay the [235]U content in radioactive centrifuge sluge from the fluorinel dissolution process[71]. It is important to note here that a comprehensive review of NDA methods for determing burnup and/or fissile content of irradiated nuclear fuels has recently been completed[72]. This in-depth review covers all applicable NDA techniques including gamma spectroscopy, passive neutron counting, active neutron interrogation (including [252]Cf source interrogation[73]) neutron resonance absorption, reactivity and calorimetry.

A particularly interesting application of neutron coincidence counting in the area of international safeguards, is the portable High Level Neutron Coincidence Counter (HLNCC) which was developed for evaluation by the International Atomic Energy Agency[74]. The portable HLNCC, shown in Figure 4, was designed for field use by IAEA inspectors in the assay of a wide variety of plutonium samples. The term "high level" refers to the high neutron count rates (e.g., up to ~ 10[5] counts/sec) produced by large (several kg) PuO_2 or plutonium metal samples. The recently upgraded detector[75] consists of 18 [3]He proportional counters embedded in 6 polyethylene slabs which form a hexagonal well (cf., Figure 4); the complete assembly is ~ 30 cm wide by 75 cm high and weighs approximately 35 kg. The newly-developed electronics package for the IAEA HLNCC, shown in Figure 5, includes high-voltage supply, amplifiers, discriminators, shift register coincidence circuitry, and control and display circuitry[75]. With the new HLNCC a 1 kg plutonium sample can be measured to a standard deviation of 1% or better in 1000 seconds.

In addition to measuring oxide and metal samples in various configurations and containers, the HLNCC has been applied to the measurement of plutonium content in mixed oxide LWR fuel assemblies[76] and to the assay and independent verification of fuel inventories in fast critical assemblies[75,77]. The latter application represents an important safeguards area both for international and domestic safeguards, and warrants some further discussion here.

Critical assembly research facilities are used to simulate advanced reactor designs -- the largest such facilities being found in the United States, United Kingdom, Japan, Federal Republic of Germany, France, and the USSR. A typical critical facility maintains an inventory of a few hundred to a few thousand kilograms of relatively pure and highly enriched SNM distributed in many thousands of small fuel pieces. As international safeguards under INFCIRC/153 (the so-called NPT "Model Agreement") come to be applied in the large industrial

nations of Europe, in Japan, the United States and the United Kingdom, some of the larger critical facilities will soon be coming under IAEA safeguards.

The major safeguards problem is the timely verification of in-reactor inventory during periods of reactor operation. This will require a judicious application of measurement techniques and careful design of statistical sampling plans to permit the incorporation of routine inspection and verification activities into normal facility operations. Technical implementation, as presently foreseen[23] will employ rapid NDA techniques to measure collectively the fuel pieces contained in reactor fuel drawers and in vault storage canisters, and to perform integral measurements of reactivity on the entire reactor inventory (in suitable reference configuration) -- reactivity measurements being particularly sensitive to small discrepancies in the reactor inventory. The portable HLNCC has been evaluated, calibrated and deployed in IAEA inspections at a large fast critical facility[75,77]. Since neutron coincidence counting determines the ^{240}Pu content, supplementary gamma measurements of isotopic ratios are needed to provide an independent verification of fissile content. Figure 6 shows a ZPPR critical assembly fuel drawer being assayed using a combination of the HLNCC (in horizontal configuration) for coincidence counting and an IAEA intrinsic germanium detector with its collimator for measurement of plutonium isotopic gamma-line ratios. This combination of neutron plus gamma ray techniques for inventory verification in fast critical assemblies has the potential for extensive application in IAEA international safeguards inspection. Integration of such state-of-the-art rapid measurement and verification technology with modern containment and surveillance measures[78] and their practical application to critical facilities should provide a high level of assurance that diversion of significant quantities of SNM can be detected on a timely basis.

VIII. INTERNATIONAL SAFEGUARDS AND THE FUTURE

There is today an increasing awareness and appreciation of the global nature of the safeguards problem and the vital importance of effective international (IAEA) safeguards -- as well as the various individual nations' safeguards systems that are the essential "building blocks" of an effective international safeguards system*. Clearly the overall goal for safeguarding the worldwide nuclear industry is an ensemble of effective national systems meeting certain broad consensus standards -- the whole ensemble functioning under an overlay of truly effective international safeguards inspection and verification. As we are all acutely aware, we're a long way from this important goal, and its achievement will clearly require much dedication and hard work on the part of the IAEA and individual nations -- including both supplier and recipient nations.

In describing herein certain elements of the U. S. Safeguards R&D program, I've tried to convey something of the thrust of current U. S. safeguards technology in respect to both international and domestic safeguards. It is important to note that although most basic technology developments have potential application in both international and national safeguards, the necessary follow-through phases of engineering design, test and field evaluation may result in significantly different final hardware products, calibration procedures, and deployment methods.

An important part of the overall safeguards R&D effort in the United States is the development of technology and equipment in response to specific needs of the International Atomic Energy Agency. The IAEA performs certain

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*It should be noted that "safeguards", as used by the IAEA, implies international safeguards; to avoid any possible confusion, we have used the qualifying adjectives, "international" or "domestic" (or "national") when a distinction between the two levels of safeguards is desired.

essential functions in carrying out its mandate of independent verification of each facility subject to IAEA safeguards within a given State. These functions include review of facility design, operating records and reports, and verification of nuclear materials accounting procedures and records, including development of inspection sampling plans and independent verification by direct assay of nuclear materials. Instrumentation needs for the latter function are many, but for present pragmatic purposes, these needs can be lumped into two time categories: (1) the "here and now" needs -- e.g., portable or transportable NDA, or other, measurement equipment for reliable assay and verification in the field, and (2) the "coming attractions" needs -- e.g., methods, instruments and techniques for independent verification of safeguards effectiveness of various types of advanced in-plant materials accounting and control systems (i.e., as given in Table 1).

In response to the category 1, "here and now" needs, portable NDA instruments and technologies specifically required by IAEA inspectors for field use are being developed, evaluated, and implemented* in cooperative programs between the IAEA and different member States. One such program, in the case of the United States for example, is the special technical assistance program which is coordinated by the International Safeguards Project Office (ISPO) at Brookhaven National Laboratory and participated in by several U.S. DOE laboratories including Los Alamos Scientific Laboratory, Sandia Laboratories, Argonne National Laboratory, Pacific Northwest Laboratory, and Brookhaven National Laboratory. The major task areas of the program[78] are directed at six functions of IAEA safeguards activity: (1). measurement technology, (2). training, (3). system studies, (4). information processing, (5). surveillance and containment, and (6). support for field operations. U. S. technical experts and consultants are also provided on a cost-free basis under individual contracts with the IAEA.

The category 2, "coming attractions" needs are closely linked with coming dramatic changes in international safeguards implementation over the next few years:

(1). As more facilities come under IAEA safeguards, there will be a great increase in the sheer volume of information that must be gathered, assimilated and analyzed.

(2). Pursuant to "NPT safeguards" concluded under INFCIRC/153, new types of large, high throughput facilities located in large industrial nations will come under IAEA safeguards for the first time. Such key fuel cycle facilities include isotope separation plants, spent fuel reprocessing plants, conversion and fuel fabrication plants producing mixed oxide fuel for power reactors, fabrication plants for highly enriched uranium fuel for research reactors, and large critical assembly facilities.

(3). With the rapid increase in the number and size of facilities under international safeguards, the IAEA will be required to deal with complete nuclear fuel cycles within individual nations, or closely coupled international/regional groups of nations with relatively less information available on nuclear materials transfers from supplier to recipient.

(4). For a host of technical and economic reasons, including operational efficiency, quality and process control, radiological and criticality safety -- not to mention the need to meet increasingly

*A notable case in point is the IAEA High Level Neutron Coincidence Counter[75] (HLNCC, described in Section VII. above) which has recently been evaluated and deployed by the IAEA inspectorate[75-77].

stringent safeguards and security requirements -- large scale facilities of the future will employ timely, on-line material measurement and accounting systems together with automated processing, remote handling equipment etc., to the maximum extent practicable. Thus it is essential that appropriate methods and techniques be developed for effective inspection and verification by IAEA (and national) inspectors.

Given the growing trend toward automation and increased sophistication in nuclear material measurement, processing and handling systems in today's competitive worldwide nuclear industry, the challenge of effectively safeguarding that industry is clear. But along with the challenge comes an important new opportunity, inasmuch as advanced materials accountancy systems can, in fact, provide far more incisive knowledge (in both time and space) of plant inventory than has ever been available in the past. This knowledge must, of course, be fully available to the inspector as well as the plant operator. To further strengthen independent verification capabilities, new techniques and procedures are being investigated to enable the inspector to carry out necessary independent calibration and measurement control functions on various assay instruments, material flows, process operations, etc. Also in the large automated fuel cycle facilities envisaged for the future, there would be minimal personnel access to hazardous in-process material (e.g., plutonium), and this strict containment feature will certainly provide an added measure of protection against theft or diversion of SNM. Furthermore, full-time resident inspection is anticipated in the large-scale regional fuel cycle plants of the future, thus giving the inspector more opportunity to gain better understanding and familiarity with plant operations, material accounting and control.

Whatever fuel cycles, or "mix" of fuel cycles, are to be pursued in various countries, groups of countries, or regions of the world, the nuclear safeguards community can, and I believe will, continue to meet the challenge of developing and implementing effective safeguards for the fuel cycle facilites of the future.

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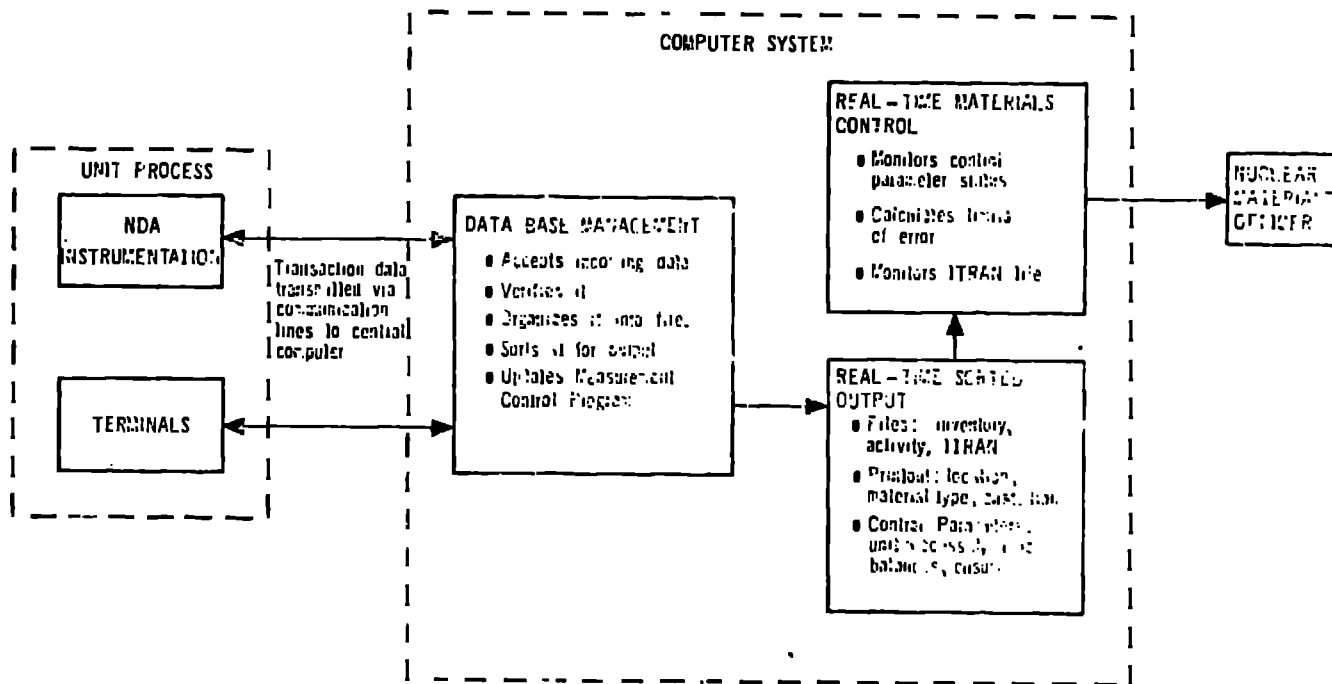
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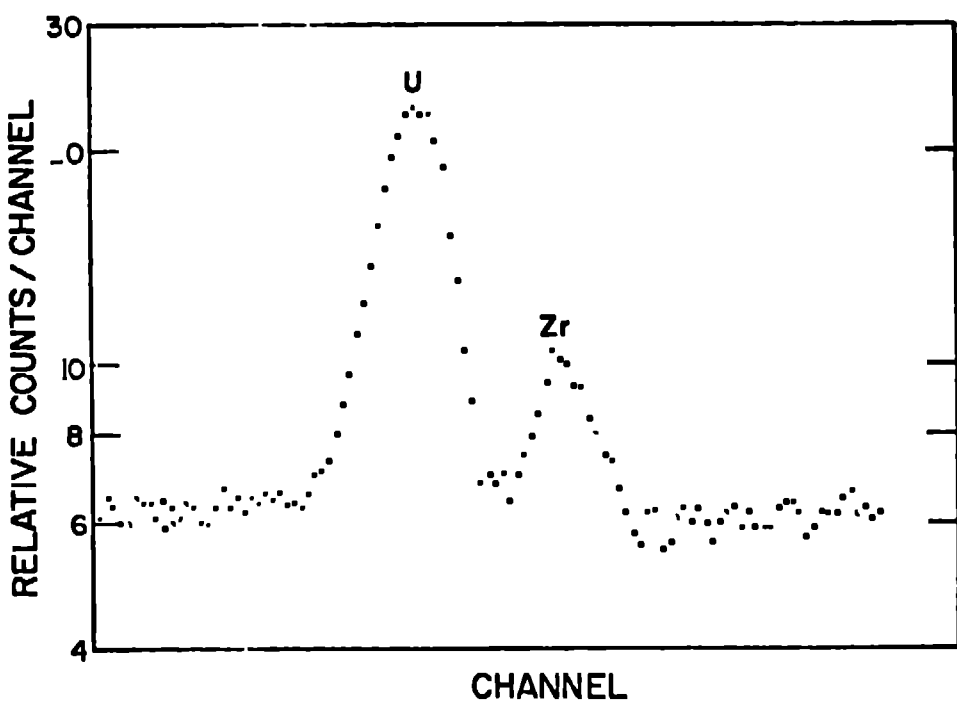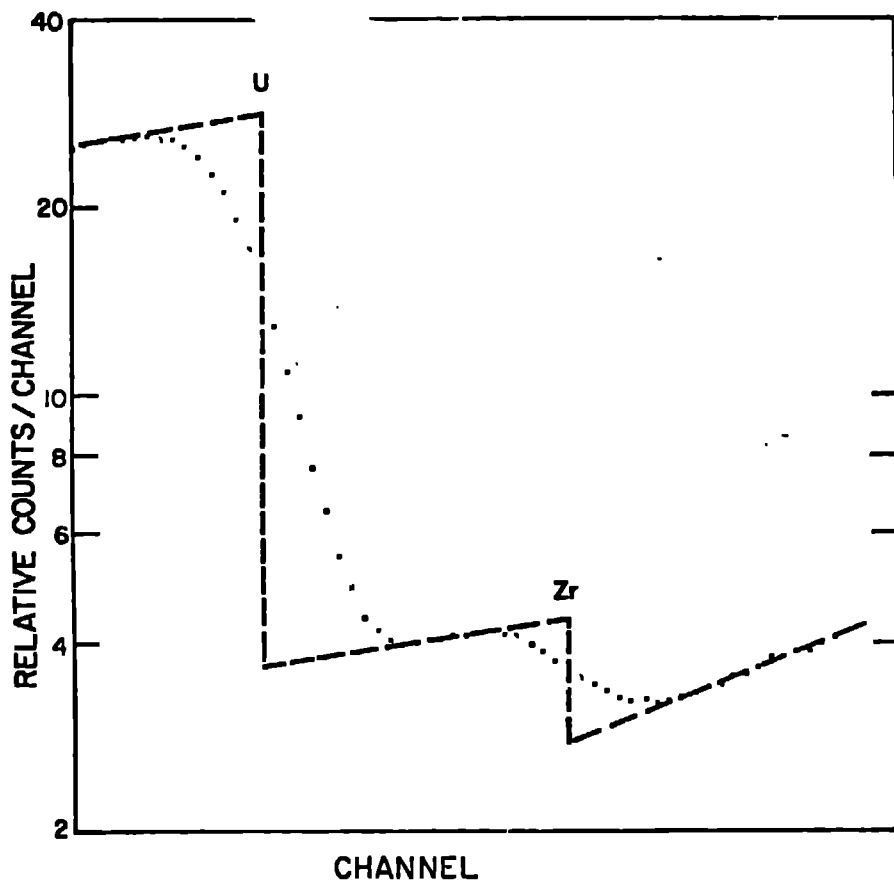
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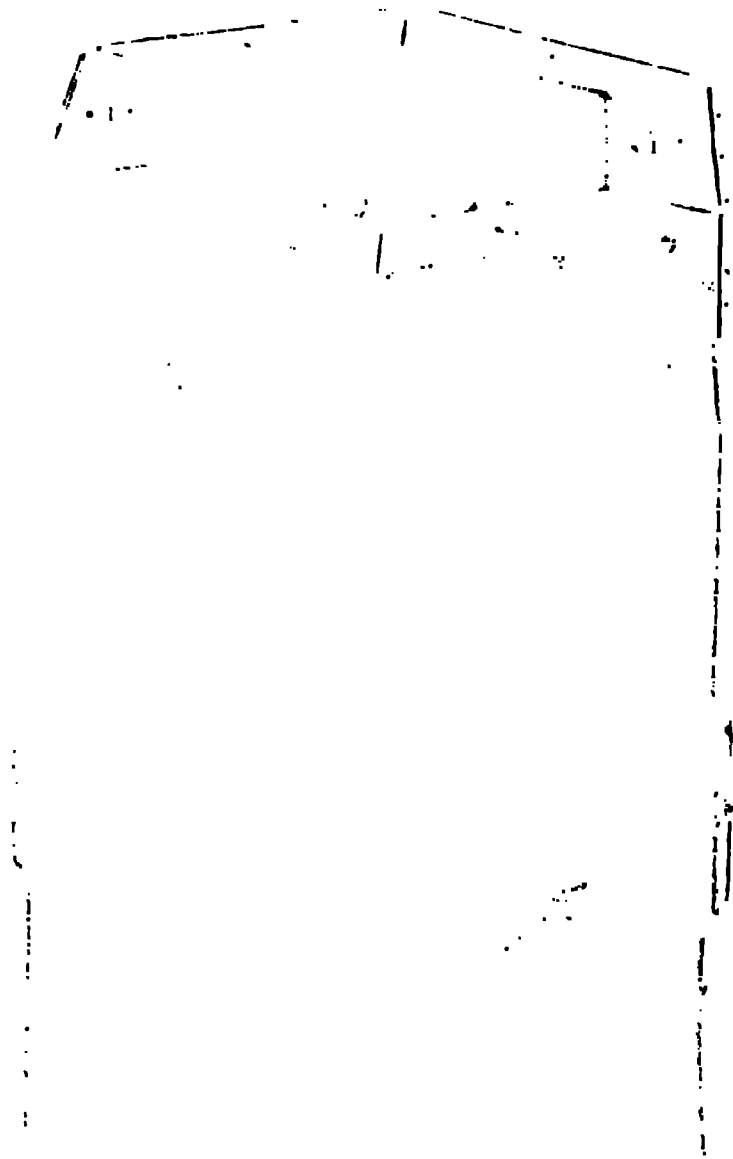69. T. W. Crane, LASL Report LA 6849-PR, pp. 20-21, (1977).

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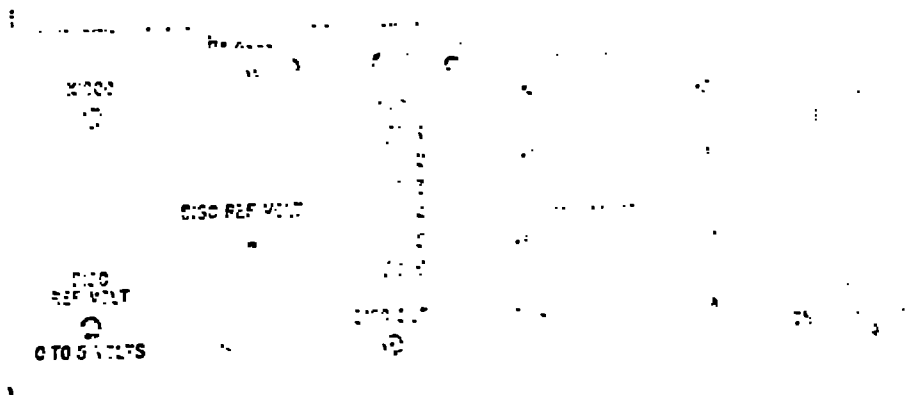
FIGURE CAPTIONS

Fig. 1. DYMAC Instrumentation for the Plutonium Recycle Wing of the
New Plutonium Facility (TA-55) at Los Alamos.

Fig. 2. DYMAC Information Handling System.

Fig. 3. Absorption Edge Densitometry of Multiple SNM Solutions.
  a (upper plot). Continuous x-ray spectrum transmitted through
                   a solution containing uranium and zirconium.
                   Data are straight-line extrapolated to the absorption
                   edges to determine the transmission ratios at the
                   edges.
  b (lower plot). The difference in the log of the measured transmission
                   spectrum (in a above) for a fixed-energy increment as
                   a function of energy.

Fig. 4. IAEA Portable High Level Neutron Coincidence Counter (HLNCC) for
Assay of High-Mass Plutonium Samples.

Fig. 5. Electronics Package for the IAEA HLNCC, Including High-Voltage
Supply, Six Amplifiers, Discriminators, and Shift-Register Coincidence
Circuit.

Fig. 6. Measurement Setup for a ZPPR Drawer, Showing the HLNCC for Neutron
Coincidence Counting and the IAEA Intrinsic Germanium Detector with
its Collimator for Measurement of Plutonium Gamma-Ray Line Ratios.

LEGEND

EVAPORATORS   AMERICIUM   OXALATE PRECIPITATION & CALCINATION

CONVEYOR TUNNEL

METAL PRECIPITATION   ELECTROREFINING

Pu NITRATE STORAGE

SAND & CRUCIBLE DISSOLUTION

SAND & CRUCIBLE FILTRATION & ION EXCHANGE

INCINERATION

IMPURE MATERIALS DISSOLUTION

IMPURE MATERIALS LEACHING

IMPURE MATERIALS FUSION, DISSOLUTION & PRECIPITATION

PEROXIDE PRECIPITATION, HYDROFLUORINATION & METAL REDUCTION

Pu NITRATE STORAGE

SPECIAL ISOTOPES

ION EXCHANGE, IMPURE Pu

RECYCLE WING

COMPUTER SYSTEM

UNIT PROCESS

NDA
INSTRUMENTATION

TERMINALS

Transaction data
transmitted via
communication
lines to central
computer

DATA BASE MANAGEMENT

• Accepts incoming data
• Verifies it
• Organizes it into files
• Sorts it for output
• Updates Measurement
  Control Program

REAL-TIME MATERIALS
CONTROL

• Monitors control
  parameter states
• Calculates limits
  of error
• Monitors ITRAN life

REAL-TIME SCALED
OUTPUT

• Files: inventory,
  activity, ITRAN
• Printout: location,
  material type, transaction
• Control Parameters,
  unit process logic and
  balances, closure

NUCLEAR
MATERIAL
OFFICER