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TITLE **OPERATION OF AUTOMATED NDA INSTRUMENTS FOR IN-LINE HEU ACCOUNTING AT Y-12**

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OPERATION OF AUTOMATED NDA INSTRUMENTS FOR IN-LINE
HEU ACCOUNTING AT Y-12[a]

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ABSTRACT

Two automated nondestructive assay instruments
developed at Los Alamos in support of nuclear
materials accounting reeds are currently oper-
ating in-line at the Y-12 Plant[b] for recovery of
highly enriched uranium. One instrument pro-
vides the HEU inventory in the secondary solvent
extraction system, and the other monitors HEU
concentration in the secondary intermediate
evaporator. Both instruments were installed in
December 1982. Operational evaluation of these
instruments has been a joint effort of Y-12 and
Los Alamos. This has included comparison of the
solvent extraction system inventories with di-
rect measurement performed on the dumped solu-
tion components of the solvent extraction sys-
te , as well as comparisons of concentration
assay results with the external assays of sam-
ples withdrawn from the process. The function,
design, and preliminary results of the opera-
tional evaluation are reported.

1. INTRODUCTION

Two automated prototype nondestructive as-
say (NDA) instruments are undergoing operational
evaluation at the Oak Ridge Y-12 Facility for
recovery of highly enriched uranium (HEU). The
instruments were designed to measure uranium
concentrations in line and to automatically de-
duce HEU inventories or provide operational in-
formation for the purposes of process control.
Each instrument uses a computer-automated data
acquisition system to obtain and analyze the
NaI(Tl) gamma-ray spectra characteristic of
^{235}U, the primary source of radiation in these
solutions. The instruments were designed[1] by
the Los Alamos National Laboratory Safeguards
Assay Group based on in-plant NDA measurements
using portable equipment[2] and on recommenda-

tions from Y-12 personnel. Operational evalu-
ation is a joint effort of Y-12 and Los Alamos.

Two nuclear materials accounting neeus
identified with high priority by Y-12 establish
the purposes for design of these prototype in-
struments. One of these needs is a timely and
reliable method, immune to large biases, for
determination of HEU inventory in the secondary
solvent extraction system. The second need is
the ability to measure and/or minimize the HEU
holdup (due to overconcentration and precipita-
tion of solids) in the secondary intermediate
evaporator that precedes the secondary solvent
extraction system in the recovery process. It
was essential that the methods developed to
satisfy these needs, as well as the development
and testing of the methods, be compatible with
production routines and schedules.

The solvent extraction system inventory
need is addressed by the EUREKA (Enriched URa-
nium Extraction Koluun Assay) instrument. EU-
REKA combines measured concentrations of uranium
in the solvent extraction columns (during pulsed
operation just before shutdown for inventory
and in the static mode after shutdown) with a
library of process design information to deter-
mine (via deduced concentration profiles in the
columns) the HEU inventories in the external
plumbing and in the columns. The intermediate
evaporator holdup problem is addressed by an
instrument that monitors uranium concentration
during operation. More timely manual control
(by the operator) of feed and product flow rates
is possible with near-real-time concentration
information made available by this instrument.
Although function and design differ substantial-
ly in the two instruments, the methods of radi-
ation detection, uranium concentration assay,
and measurement control are common to both.

Operational evaluation of the two prototype
instruments began in December 1982. These two
independent evaluations are still in progress.
This paper[c] emphasizes the design and functional
aspects of these instruments and evaluates the
measurement control techniques. The preliminary
results are also reported.

II. INSTRUMENT FUNCTION AND DESIGN

A. EUREKA Function and Design

The Y-12 facility for recovery of highly enriched uranium operates two identical parallel solvent extraction systems for secondary purification of uranium in concentrated (200 g/L) acid (4 $\underline{M}$ HNO$_3$) solution. Figure 1 is a schematic illustration of one of these systems consisting of three pulsed solvent extraction columns, each 10 m tall by 10 cm in diam with glass (upper one-third) and stainless steel sections.[3] The glass sections are inactive, and the (active) stainless steel sections are fitted at 2.5-cm intervals with 1.6-mm-thick stainless steel sieve plates for phase dispersion. Uranium is extracted from the acid solutions by the low-density organic phase in the extraction column, and is stripped from the organic phase into the low-acid aqueous phase in the backwash column. The organic products of the extraction and strip columns are typically saturated ($\sim$120 g/L) with uranium. The raffinate stream uranium concentration is a few grams per liter, and the aqueous product concentration is approximately 100 g/L. Except for the aqueous feed and product streams and raffinate streams, each system operates in a closed loop. During the monthly shutdown, the aqueous feed and product storage tanks and the raffinate storage tanks are sampled and assayed for inventory. A separate inventory determination, excluding the storage tanks, is required for the HEU (approximately 25 kg per system) in each of the two solvent extraction systems. This inventory is presently performed on one of the two parallel systems by the EUREKA instrument.

Figure 2 is a conceptual illustration of EUREKA. Six shielded NaI(Tl) detectors, mounted simultaneously along the solution-bearing length (9 m) of any one of the three columns of the secondary solvent extraction system are used to determine the uranium concentration at the six vertical locations on each column. The six detectors mount on brackets clamped to (or adjacent to) each column in a fixed position. The detectors are moved from column to column to perform the assays in count times of 600 s.
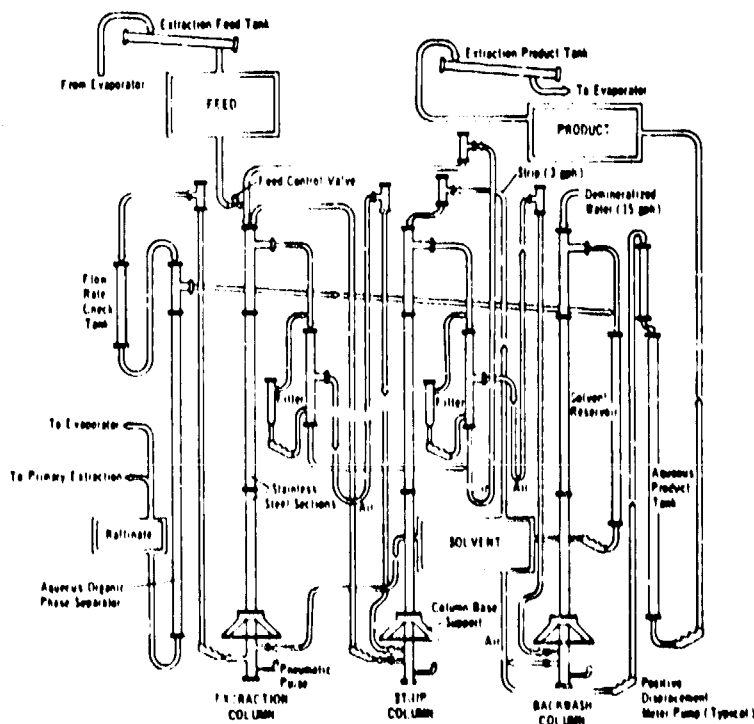

Fig. 1. Schematic illustration of one of two secondary solvent extraction systems for purification of HEU at Y-12.

OPERATIONS AREA   **CONTROL AREA**
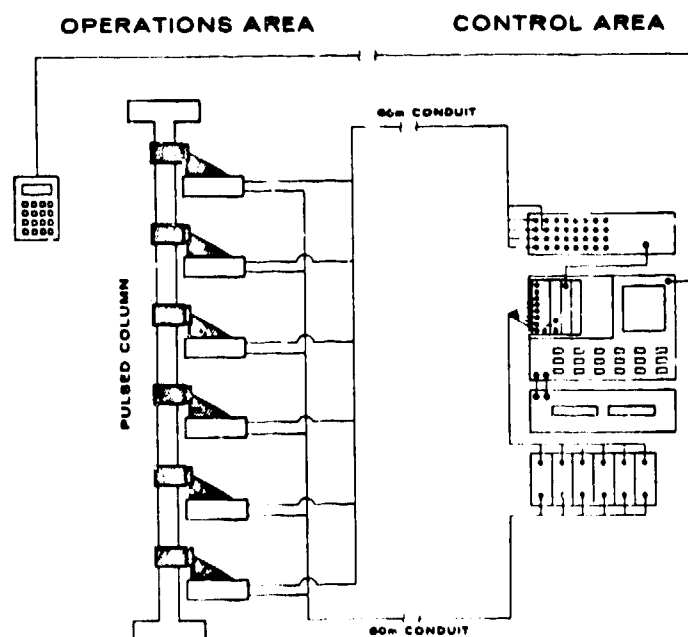
Fig. 2. The six shielded detectors, mounted on one of
the solvent extraction columns in the operating area
(left), and the electronics in the remote, controlled-
environment area (right) are shown schematically to il-
lustrate the EUREKA design. The electronics package in-
cludes (bottom to top) six stabilized amplifiers, a dual
floppy disk drive, a computer-based multichannel analyzer
with CAMAC minicrate (containing multiplexer-router, ADC,
high-voltage supply interface, and clock), and a program-
mable high-voltage power supply. The portable terminal
for control of the assay in the operations area is also
shown.

Column inventories are determined from the meas-
ured concentrations by construction of a con-
centration profile for each column in the ver-
tical dimension. The column inventory is the
numerical integral of the product of column
concentration and volume element.

All electronics components downstream of
the detectors reside in a remote, controlled-
environment location at Y-12, as illustrated in
Figure 2. Figure 3 is a photograph of these
electronics assembled before shipment to Y-12,
and Figure 4 shows the electronics installed at
Y-12. The six linear amplifier outputs are
multiplexed into a single analog-to-digital
converter (ADC) and routed for separate storage
in the computer-based multichannel analyser.
Analog gain stabilization is employed, and the

computer maintains control of the high voltage
on each of the detectors. The CRT display auto-
matically updates any of the six pulse-height
spectra in real time (during data acquisition)
and provides the option to examine any spectrum
in detail after acquisition. The data are auto-
matically analyzed, and the results (uranium
concentrations and inventory) are printed at the
end of each analysis.

Figure 5 is a photograph of the shielded
detector assemblies prior to shipment to Y-12.
Figure 6 is a photograph taken at the Y-12 so-
lution recovery area showing a shielded detector
mounted on a stainless steel section of a sol-
vent extraction column.

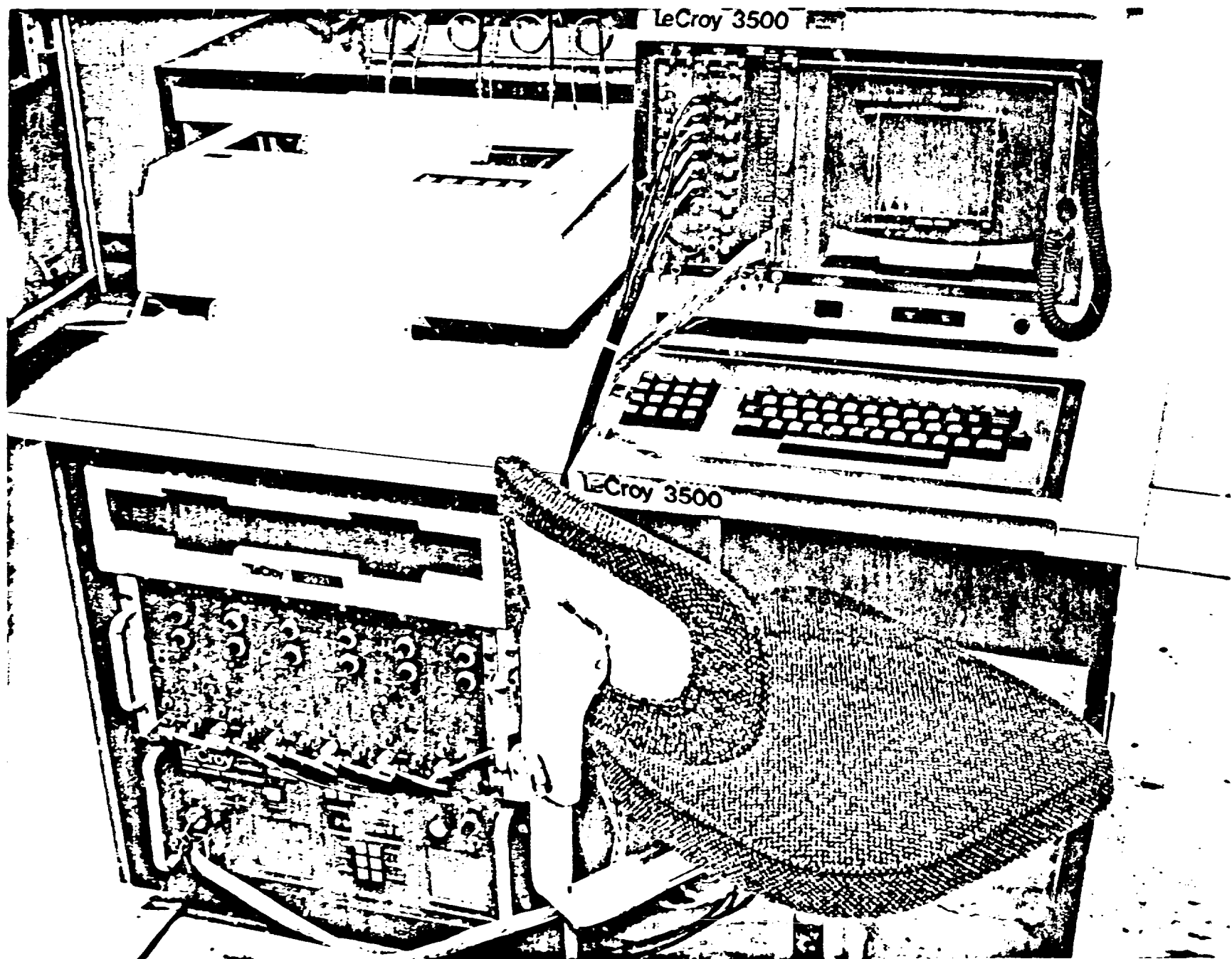Fig. 3. Electronics for EUREKA, assembled prior to shipment to Y-12. This equipment, located in the controlled-environment area at the Y-12 HEU recovery facility, consists of (left, bottom to top) the programmable high-voltage power supply, the stabilized amplifiers, the dual floppy disk unit, and the hardcopy unit, as well as (right) the programmable multichannel analyzer with CAMAC minicrate.

Fig. 4. EUREKA electronics installed in the controlled-environment area at Y-12. A shielded detector is held in the foreground of the photograph. The terminal for control of the assay in the operations area is shown in front of the hardcopy unit.

Fig. 5. Shielded detectors for EUREKA, assembled prior to shipment to Y-12. The uranium (working standard) disks, mounted on the tungsten holders, are shown inserted in the six leftmost collimator shields. The portable terminal for control of the assay in the operations area is in the background.

The most accurate and precise results for uranium concentration can be achieved by measurements of single-phase solutions in the static columns. This is accomplished at the normal inventory period when the columns have been shut down and pulsed to disengage phases. However, approximately 20% of the system inventory resides in the plumbing external to the solvent extraction columns. Each plumbing line either feeds or is fed by solution in the column. Therefore, column concentration profiles can also be used to deduce uranium concentrations in solutions that reside in the solution feed and drain lines. Measurements to obtain concentration profiles of the static columns are performed after shutdown and pulsing of the columns so that the memory of the concentration in the external plumbing is effectively erased from the static profiles. However, profiles obtained from measurements of the columns during pulsed operation (just before shutdown for inventory) provide the concentration information necessary to determine the inventory in the external plumbing. Therefore the columns are measured first during pulsed operation just before the monthly shutdown for inventory and again following shutdown and pulsing for phase separation.

Fig. 4

Jan 5

feed, a shielded NaI(Tl) detector was installed on the evaporator return loop, and an automated instrument was designed to provide a near-real-time readout of uranium concentration in the evaporator solutions.

Figure 7 is a schematic illustration of the monitor installed on the Y-12 secondary intermediate evaporator. The electronics components, located approximately 5 m from the evaporator, are enclosed in an environmental cabinet through which instrument air flows continually. The small, programmable data acquisition and control unit automates the assay and the monitor readout. The readout panel displays the concentration result on a numerical LED display and in an array of colored lights (visible to the operator controlling the evaporator solution flow

Fig. 6. Operator at the Y-12 facility for recovery of HEU is shown positioning a EURFKA detector on the extraction column of the secondary solvent extraction system.

The assay system has been designed to automate (under computer control) all operations except for positioning the detectors, performing the appropriate sequence of process operations during the assay, and providing input of process information used by the assay. Control of the assay is accomplished either remotely at the main terminal (in the controlled-environment location) or at a portable terminal in the process area (refer to Figs. 4 and 5) so that the process operator can perform the assay. Control of the assay at the portable terminal enables use of this equipment for process monitoring applications.

B. Evaporator Monitor Function and Design
Concentration of uranium in solution for feed into the Y-12 secondary solvent extraction system is carried out by the secondary intermediate evaporator. This is a recirculating, steam-jacketed unit, approximately 6 m tall, with an isolated 9-cm-diam stainless steel pipe for solution return to the steam-jacketed section. Control of the evaporator is accomplished by manual operation of feed and product flow rates based on monitoring of the specific gravity of solutions withdrawn from the evaporator during operation. This time-consuming process gives rise to frequent upsets in operation. Furthermore, the specific gravity is strongly dependent on acid molarity in these highly acidic solutions. To avoid overconcentrating (and precipitating) uranium and to assure the desired uranium concentration for solvent extraction

Fig. 7. Schematic illustration of uranium concentration monitor installed on the Y-12 secondary intermediate evaporator. The shielded detector is shown mounted on the evaporator solution return loop. The electronics package (right), located 5 m from the evaporator, consists of (left to right) a stabilized amplifier, high-voltage supply, ADC, and programmable data acquisition and control (PDAC) unit. The LED display unit is also shown. The capability for automatic control of valves by the PDAC unit, although not presently implemented, is illustrated here.

Fig. 6

rates) that indicate concentration range. Read-out updates occur, typically, at 30-s intervals. The electronics cabinet and the shielded NaI(Tl) detector are shown, installed at Y-12, in Fig. 8.

C. Uranium Concentration Assay

The counting geometry for assay of uranium concentration in the solutions in the secondary solvent extraction system is illustrated in drawings of the radial and longitudinal column cross sections, Figs. 9 and 10, respectively. A similar geometry applies to the assay of solutions in the evaporator return loop except that the thickness and inner diameter of the stainless steel pipe are 0.55 and 8.9 cm, respectively, and there are no sieve plates and stator rods to consider.

The assay signature for the uranium concentration measurement is the 186-keV gamma ray of ^{235}U. Therefore, the accuracy of the assay relies on a known value of the uranium enrichment in these solutions.

Because solvent densities in the secondary solvent extraction system can vary by ∿25% about unity, solution self-attenuation effects are solvent dependent. Thus calibrations were calculated for each solvent category. The calculation for the low-acid, aqueous solutions within the stainless steel portions of the solvent extraction columns was verified empirically.

The calculation integrates the 186-keV activity (at the cone apex) from a truncated

Fig. 8. The equipment for the uranium concentration monitor for the secondary intermediate evaporator is shown installed at Y-12. (a) The electronics components in the environmental enclosure are (left to right) the programmable data acquisition and control unit, the high-voltage power supply, the stabilized amplifier, and the ADC. Above the environmental cabinet is the LED display unit. (b) The shielded NaI(Tl) detector is mounted on the return loop of the secondary intermediate evaporator.
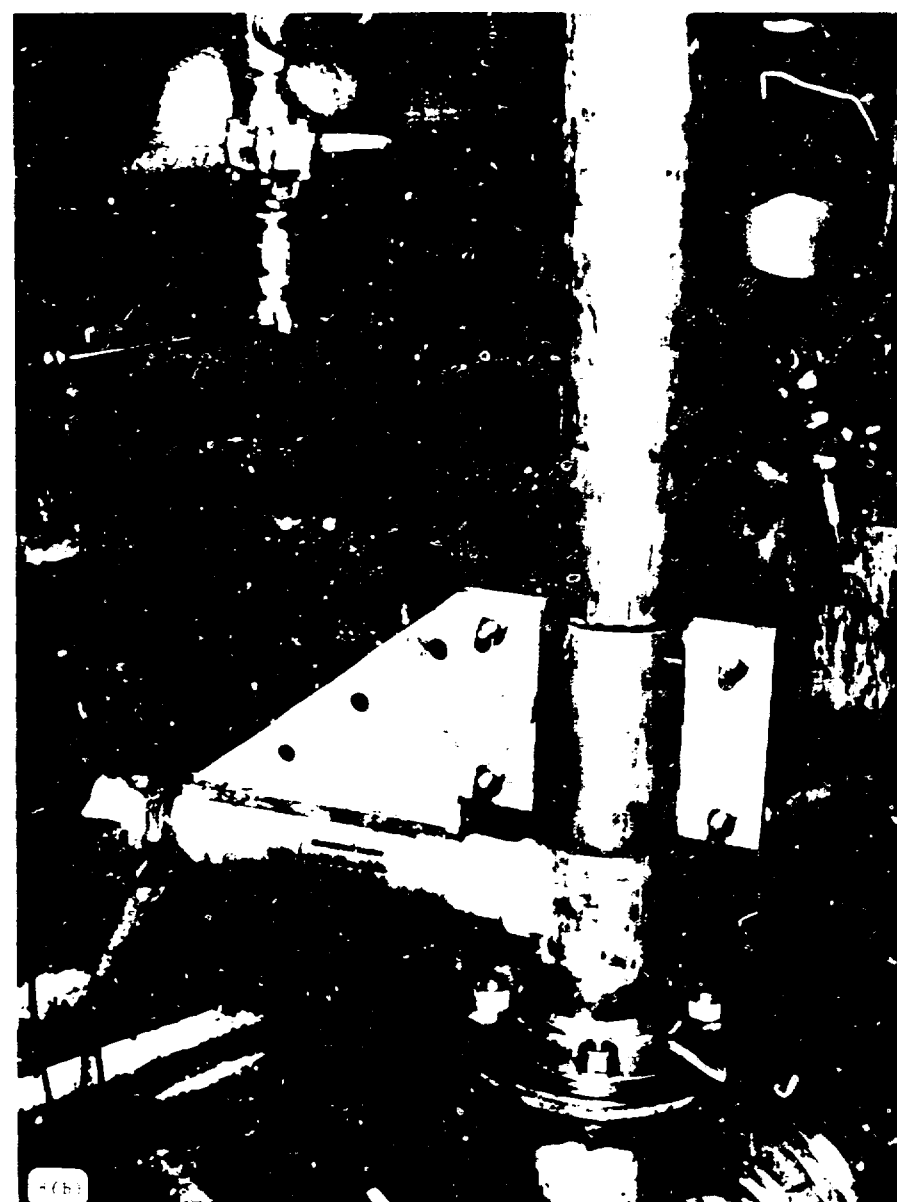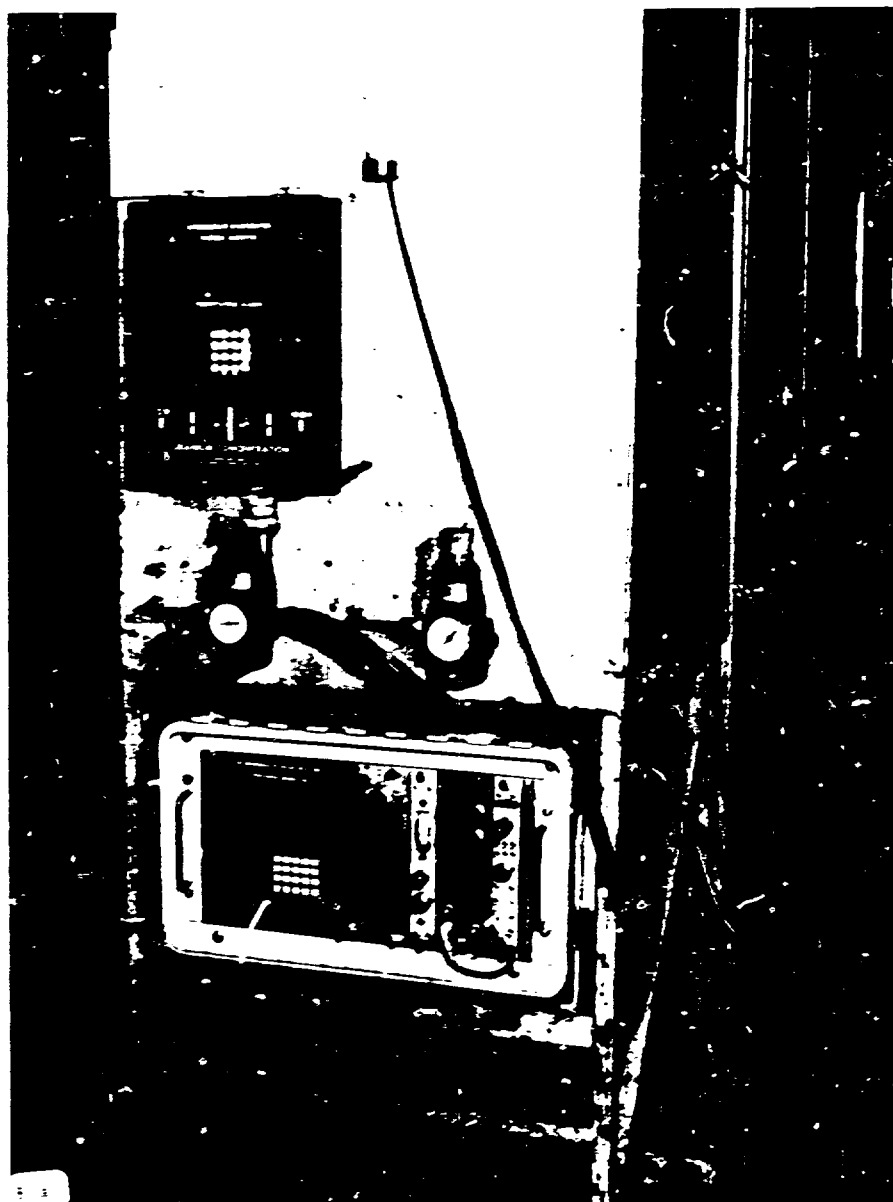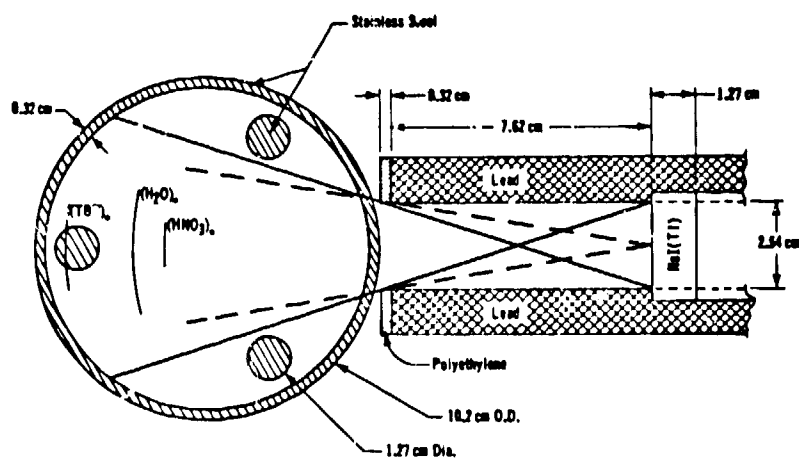
Fig. 9. Drawing to scale of a radial cross section of a stain-
less steel section of a secondary solvent extraction column in
the active region. The detector position with respect to the
stainless steel stator rods corresponds to the actual positions
of the EUREKA detectors on the solvent extraction columns. The
curves labeled $(H_2O)_o$, $(HNO_3)_o$, and $(TBP)_o$ mark one mean free
path for 186-keV gamma rays in pure water, nitric acid (4 $\underline{M}$),
and 30% tributyl phosphate in kerosene.

Fig. 10. Drawing to scale of a longitudinal cross section of
a stainless steel section of a secondary solvent extraction
column in the active region. The detector position with re-
spect to the sieve plates corresponds to the maximum count-
rate configuration for 186-keV gamma rays. Refer to the cap-
tion of Fig. 9 for definitions.

right-circular cone-shaped source of uniform activity. It includes the attenuation effects of the sample and external absorbers, and 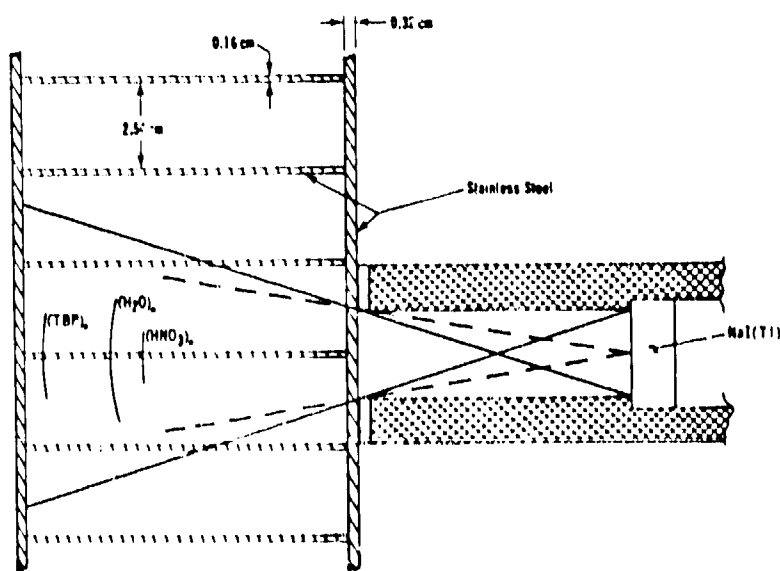incorporates a reduced scattering attenuation to account for gamma rays that scatter but are still detected within the resolution limits of the 186-keV photopeak. The calculation does not include the effects of the plates and rods.

The measurements performed to verify the calculation required fabrication of a stainless steel sample cell (Fig. 11) to reproduce the construction and configuration of the Y-12 secondary solvent extraction columns. The lower half of the 50-cm-long by 10-cm-diam cell contained appropriately positioned plates and rods designed to match the sieve plates and stator rods that exist in the active regions of the solvent extraction columns. Separate measurements were performed with a reference detector positioned on the upper and the lower half of the cell. To eliminate the effects of count-rate losses on the assay result, the measured 186-keV count rate is normalized to that for the 60-keV photopeak produced by a 0.5-µCi ^{241}Am

source attached to the detector in fixed geometry. The normalization factor differs for each detector, due primarily to differences in the ^{241}Am source strengths. Measurements were performed using five well-characterized HEU solutions with uranium concentrations of 7, 59, 129, 178, and 258 g/L.

The absolute 186-keV count rates measured with the reference detector positioned on the upper half of the cell are within 1% of the calculated value with a standard deviation of 0.5%. The corresponding relative count rates (the 186-keV rate divided by the 60-keV rate) and the calculated calibration normalized to these results are the large dots and the solid line, respectively, in the upper portion of Fig. 12. The ratios of the measured to the calculated values are shown in the lower portion of Fig. 12. The relative standard deviation in this ratio for the five samples is 0.7%.

The relative count rates measured with the reference detector positioned on the lower half of the cell are also plotted in Fig. 12. These include results obtained with the detector positioned between two rods (crosses) as illustrated in Fig. 9 and results obtained with the detector positioned directly in front of a rod (triangles). The measured effects (relative to results obtained on the upper portion of the cell) of the sieve plates are plotted in Fig. 13 as a fractional decrease in the 186- to 60-keV ratio versus uranium concentration. The measured effects of the stator rods are plotted in Fig. 14.

The large effects (approximately 20% or more) of the stator rods on the measured 186-keV count rates are avoided in the EUREKA assays by positioning the detectors between the rods on the active regions of the solvent extraction columns. The smaller effects of the sieve plates, represented by the quadratic fit to the data in Fig. 13, are incorporated in the EUREKA assays of solutions in the active portions of the columns by applying this empirical correction to the measured count rates in an iterative manner. The error bars on the data in Fig. 13 indicate the high and low results obtained from measurements with the collimator centered on a plate and between two plates, respectively. The positioning of EUREKA detectors on the active portions of the column is random with respect to the relative positions of the plates in relation to the collimator. This incorporates a small uncertainty into the concentration assay.

A separate calculation of count rate versus uranium concentration is performed for each

CROP

Fig 11
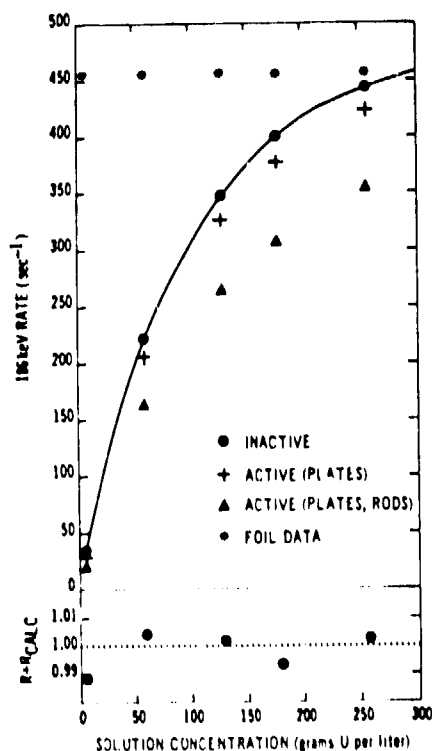
Fig. 12. Relative net photopeak
areas (186-keV area divided by
60-keV area) measured with ref-
erence detector mounted in dif-
ferent locations on the standard
stainless steel sample cell.
Measurements were performed with
five different reference solu-
tions of highly enriched uranium
(top). The solid curve is the
calculation that applies to the
measurements in the inactive (no
plates) region (large dots). The
ratios of these points to the
calculated curve are shown at the
bottom. A measurement control
data point obtained during each
set of solution assays with the
reference uranium disk inserted
in the collimator is also plotted
(asterisks). Measurements in the
active region are indicated by
crosses (plates only) and trian-
gles (plates and rods).
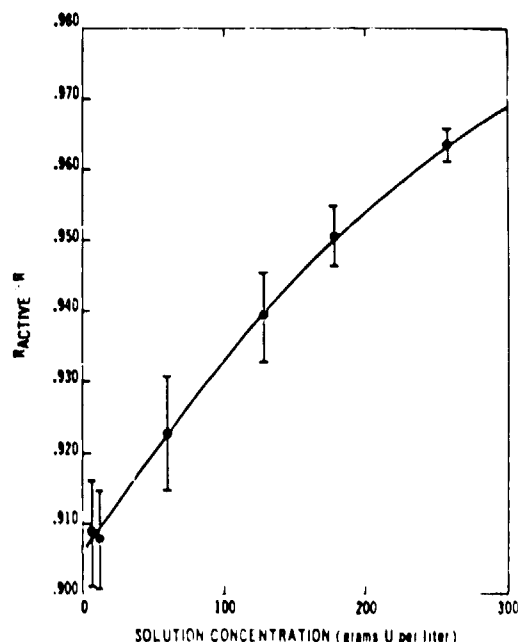
Fig. 13. The effect of the sieve plates
on the 186-keV count rate is illustrated
by the data points, which are the ratios
of the crosses to the large-dot data
points of Fig. 12, plotted versus ref-
erence solution concentration. The ac-
tual data points shown are each an av-
erage of two results: one with the de-
tector collimator positioned as in Fig.
10, and one with the detector collimator
positioned between two sieve plates.
The error bar limits (upper and lower)
show these two results, respectively.

column dimension (including that for the evapo-
rator return loop) and for each solvent phase.
A specific, representative acid molarity is as-
sumed for each calculation. The inverse of the
calculated results (concentration versus count
rate) is fitted with a (ninth order) polynomial,
and the fit parameters are stored in the data
acquisition programs. For EUREKA, the measured
count rates for solutions with acid molarities
different from those assumed in the calculation
are corrected in a second iteration in solving
for concentration. This correction, a linear
function of acid molarity, is derived from the
numerical integrations applied over the useful
range of acid molarity.
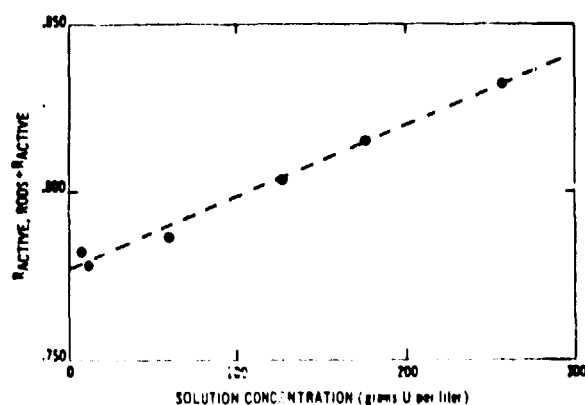
Fig. 14. The large effect of a stator rod, positioned directly in front of the detector, on the 186-keV count rate is illustrated by the data points, which are the ratios of the triangular ro the cross data points of Fig. 12, plotted versus reference solution concentration. These effects are avoided by positioning detectors with respect to rods as shown in Fig. 9.

The use of a polynomial fit to the results of the numerical integration was the simplest approach to the calibration for several reasons. The first reason is that double precision arithmetic is required to obtain the necessary sensitivity in the original numerical integration, and the polynomial fit avoids this requirement. A second reason is that an inverse form of the numerical integration was not apparent. A third reason is that the ninth-order polynomial fit to the calculated rates gives better than 0.2% agreement (in concentration) over 90% of the concentration range in which it is applied and better than 0.5% in the lowest 10% of the range.

There are several random effects that can be quantified to give an estimate of the uncertainty in the EUREKA concentration assay results. These include the effects of counting statistics, the effects of the position of the collimators with respect to the sieve plates, and the effects of the uncertain knowledge of acid molarity. The sieve plate effects are not truly random because a given detector holder, once positioned, will remain in a fixed position with respect to the plates. However, averaged over many detector positions, the effect can be treated as a random effect on the concentration result.

Table I shows the random uncertainties in the measured 186-keV count rates due to counting statistics (in 600-s counts) and positioning with respect to the sieve plates for three concentrations spanning the assay range. The net result in Table I is the quadratic sum of these two uncertainties. Table II shows the random uncertainties in the measured 186-keV count rates due to uncertain knowledge of solution acid molarities for the same concentrations. These results are shown separately for measurements of the static and pulsed columns since the uncertainties are significantly larger for the latter. Table III shows the quadratic sums of the net results from Table I and the results from Table II.

TABLE I

RELATIVE RANDOM UNCERTAINTY (1σ) IN THE 186-KEV COUNT RATE
DUE TO COUNTING STATISTICS AND SIEVE PLATES

ρ_U (g/l)	T = 600 s	PLATES	NET
10	1.1%	0.8%	1.4%
125	0.4%	0.6%	0.7%
250	0.3%	0.2%	0.4%

TABLE II

RELATIVE RANDOM UNCERTAINTY (1σ) IN THE 186-KEV COUNT RATE
DUE TO UNCERTAIN KNOWLEDGE OF ACID MOLARITY

ρ_U (g/l)	HIGH ACID		LOW ACID	
	PULSED**	STATIC	PULSED**	STATIC
10	2.8%	0.8%	1.5%	0.6%
125	2.7%	0.6%	1.1%	0.5%
250	1.7%	0.5%	0.9%	0.4%

*ASSUMES KNOWLEDGE OF HIGH ACID SPECIFIC GRAVITY TO ±1% (THAT IS, ACID MOLARITY IS KNOWN TO ±20%). ASSUMES KNOWLEDGE OF LOW ACID SPECIFIC GRAVITY TO ±2% (THAT IS, ACID MOLARITY CAN VARY BETWEEN 0 AND 2)

**ASSUMES UNCERTAINTY (2σ) IN THE AVERAGE SPECIFIC GRAVITY OF MATRIX TO BE 1/2 THE DIFFERENCE IN SPECIFIC GRAVITY OF THE AQUEOUS AND ORGANIC PHASES

TABLE III

NET RELATIVE RANDOM UNCERTAINTY (1σ) IN 186-KEV COUNT RATE

ρ_U (g/l)	HIGH ACID		LOW ACID	
	PULSED*	STATIC	PULSED*	STATIC
10	3.1%	1.6%	2.1%	1.5%
125	2.8%	0.9%	1.3%	0.9%
250	1.8%	0.7%	1.0%	0.6%

*ASSUMES UNCERTAINTY (2σ) IN THE AVERAGE SPECIFIC GRAVITY OF MATRIX TO BE 1/2 THE DIFFERENCE IN SPECIFIC GRAVITY OF THE AQUEOUS AND ORGANIC PHASES

Due to the increasing slope of the curve of uranium concentration versus 186-keV count rate with increasing concentration, the relative uncertainty in uranium concentration is much larger than the uncertainty in count rate at high concentrations. The differential relative uncertainties for the three representative concentrations are given in Table IV. The product of this quantity and the net random uncertainty in the 186-keV count rate (Table III) gives the net random uncertainty in the measured uranium concentration. These results are given in Table V.

The data of Table V can be used to predict the random uncertainty in the EUREKA inventory of a given column. This is approximately 3% for a static column and 5 or 6% for a pulsed column if no other random effects contribute.

D. Measurement Control

Measurement control applied to these inplant NDA instruments includes: systematic monitoring of photopeak centroids, photopeak widths, and count rates of all spectra acquired; regular measurements of backgrounds; and regular assays of (working "standard") enriched uranium disks. The accepted values for photopeak widths and centroids are stored in parameter files accessed by the EUREKA and evaporator monitor codes. These values are compared with the measured values. The data are automatically flagged if the deviation exceeds the prescribed value. Because analog spectrum gain stabilization is employed and because count rates are low (approximately 3000 s^{-1}) and relatively constant (since the ^{241}Am is responsible for most of the rate), it is rare that these flags are raised. Background and foil measurements are performed at intervals prescribed by the users. The schedules differ for the two instruments, but the methods are the same and the results of the background and foil measurements have the same significance for both instruments.

The method for background measurement involves insertion of a 5-mm-thick tungsten shield into a slot near the end of the collimator shield, followed by a normal assay for determination of the ratio of net peak areas (of the 186- and 60 keV photopeaks). These results are subtracted from subsequent assays of samples. Backgrounds are normally stored in the parameter file by the evaporator monitor code. This effort is completely automated (through execution of a separate background assay option) by the code, which stores a separate background result for each detector mounted on each of the three columns. (A total of 18 background results are stored.) These stored values are updated each time backgrounds are remeasured or background

spectra are read into memory from floppy diskette files. Background ratios of net peak areas are typically 1 to 5% of the ratio measured in a solution assay for which the uranium concentration is 100 g/L.

Assay of uranium disks for measurement control purposes involves insertion of a tungsten-backed 13-mm-diam disk of highly enriched uranium metal (total weight is 3 g per disk) into the same slot in the collimator that is used for the background shields. (Refer to Fig. 5 .) The uranium disks are mounted on one end of the tungsten background shields. Each detector has its own unique disk. The disk position is reproducible in that the uncertainty in the 186-keV count rate due to positioning is less than the random uncertainty due to counting statistics. The ratio of net peak areas determined in the disk assay is compared (automatically in the EUREKA code) with the nominal values for each detector. A change in the counting efficiency parameter propagates proportionally into the measured foil ratio. Therefore, the foil result has been used to monitor (and correct) the efficiency parameter for each detector.

The efficiency parameter is defined as the ratio of the normalization factor (net peak area ratio for 186- and 60-keV photopeaks) for a

TABLE IV

DIFFERENTIAL RELATIVE UNCERTAINTY IN URANIUM CONCENTRATION VERSUS THAT IN COUNT RATE

ρ_U (G/ℓ)	$\left|\delta\rho_U\ (\%)\right| / \left|\delta N_{186}\ (\%)\right|$
10	1.06
125	2.03
250	4.08

TABLE V

RELATIVE RANDOM UNCERTAINTY (1σ) IN MEASURED URANIUM CONCENTRATION

ρ_U (G/ℓ)	EXTRACTION, STRIP PULSED[a]	EXTRACTION, STRIP STATIC	BACKWASH PULSED[a]	BACKWASH STATIC
10	5.5%	1.7%	2.2%	1.6%
125	4.7%	1.8%	2.6%	1.8%
250	7.3%	2.9%	4.1%	2.4%

[a]ASSUMES UNCERTAINTY (2σ) IN THE AVERAGE SPECIFIC GRAVITY OF MATRIX TO BE 1/2 THE DIFFERENCE IN SPECIFIC GRAVITY OF THE AQUEOUS AND ORGANIC PHASES

given detector relative to that for the reference detector. This quantity is stored by each detector in the EUREKA parameter file. It can be measured directly by assay of a reference solution of uranium in the standard cell, or the directly measured values can be corrected by the changes in the disk assay results. The latter method has proven to be a simple, accurate, and essential alternative to the time-consuming effort of direct measurements of solutions in the standard cell.

The average relative standard deviation in the six disk assay results obtained (prior to each static and pulsed assay on each of the three columns) during inventories since February 1983 is 0.35% for all six detectors. This is only slightly larger than the contribution of counting statistics (0.3%) to the random uncertainty. Therefore any effects of the location on the column, the identity of the column (extraction, strip, or backwash), or the column operational status (pulsed or static), as well as effects of disk positioning in the shields, are small enough to be ignored in using the disk assay results to correct the detector efficiency parameters.

E. Results and Significance of Measurement Control by Uranium Disk Assay

Measurement control by disk or foil assay has been demonstrated previously as a method to monitor the stability of the calibration of solution assay instruments that use methods of high-resolution gamma-ray measurement based on both passive gamma-ray assay and on gamma- or x-ray transmission measurements.[4] The greater overall stability in the high-resolution systems reduces the required frequency of direct calibration even though the desired accuracy greatly exceeds that achievable by NaI(Tl) detectors. Therefore, foil assay results have been used in the past to monitor the calibration of a high-resolution gamma-ray instrument on a frequent basis. Adjustment of the high-resolution system calibration, which is required only infrequently, is accomplished directly using well-characterized reference solutions

It is not practical to rely on the availability of reference solutions to satisfy the frequent needs for recalibration of NaI(Tl) assay instruments. Furthermore, the time and effort required to apply such calibration methods frequently to multiple detectors designed to operate on-line is prohibitive. Therefore, the Y-12 instruments have relied on the disk assay results to provide the necessary calibration updates. The extensive disk assay data obtained with the EUREKA detectors (a disk assay has preceded each concentration assay performed on the

solvent extraction columns to date) illustrate the effectiveness of these methods.

Figure 15 is a plot of the efficiency factors deduced from the disk assay results versus date for each of the six EUREKA detectors during the first 8 months of operation at Y-12. With one exception, each (solid) data point since the January measurements is the average of six 600-s disk measurements. (These are the disk assays that precede the pulsed and static solution assays on each of the three columns during the monthly measurements for inventory.) The exception is a set of disk measurements performed in mid-May, concurrent with direct measurements at Y-12 of the efficiency factors (open points) for each detector using a well-characterized reference solution and the calibration cell. The dashed line indicates the efficiency factor measured for each detector by direct methods during the original calibration at Los Alamos. It was during this original calibration that the characteristic (186- to 60-keV) ratios, which have been used for comparison with all successive disk assays, were precisely determined.

The initial measurements with detector 4 just after installation of EUREKA indicated the possibility of electronic malfunction in this detector. It was subsequently disassembled, checked, and reassembled in its shield. In this process, the ^{241}Am source geometry was changed, causing a large (10%) decrease in the efficiency factor. The availability of the well-characterized foil made possible the immediate use of this detector after reassembly, without the need to recalibrate using a reference solution and the standard cell.

Large fluctuations in the efficiency factors deduced from disk assays during December through February indicated a possible shifting (sliding) of several of the NaI(Tl) detectors in the shields. Just prior to the March inventory, nylon set screws were installed to fix the proper detector positions. Since that time, only small, gradual monotonic changes in the efficiency parameters were observed. In each case (except for detector 4), a linear extrapolation of the data beginning with the March results intersects the original direct measurement (dashed line) at the y axis (late November 1982), the time of the original direct measurements.

The direct calibration measurements performed in mid-May with a 100 g/l uranium reference solution in the standard cell produced efficiency factors in excellent agreement with the disk prediction (refer to Fig. 15). The average deviation of the disk prediction from the direct
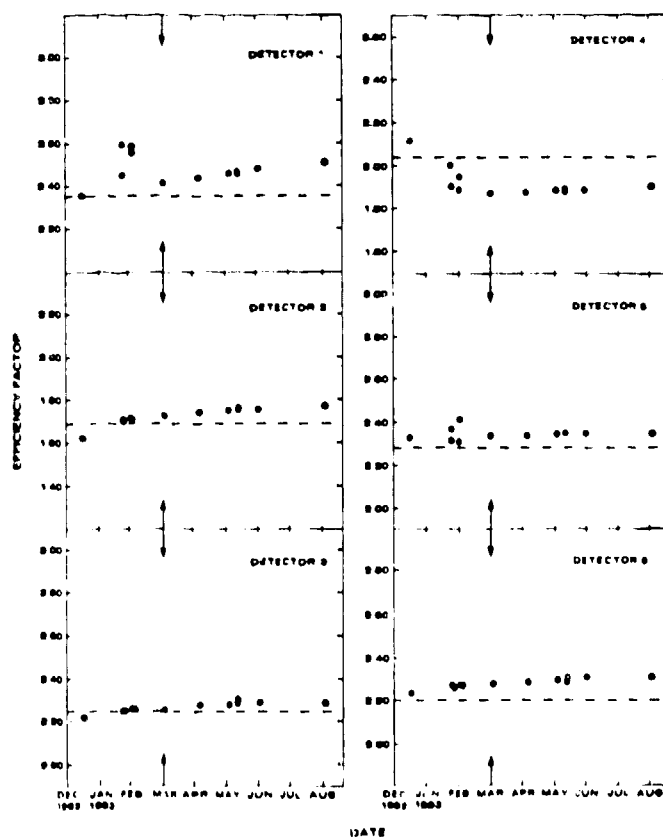
Fig. 15. The efficiency ' 'or deduced from disk assays (solid points) is plo. J versus date for the six EUREKA detectors. With the exception of the points plotted in mid-May, each point since January 1983 corresponds to the average of six disk measurements obtained prior to solution assays on each of the three columns in the pulsed and static modes during inventory. The dashed lines correspond to the efficiency factors determined by direct measurements at Los Alamos of reference solutions in the standard cell. The open circles plotted at mid-May are direct measurements performed at Y-12 using the standard cell and a uranium reference solution. (Disk measurements were also performed at this time. The disk result for detector 5 is superimposed on the direct measurement.) The vertical arrows mark the date on which the detector positions were fixed in the shields.

measurement was 0.4% and the standard deviation was 0.5%, 1σ. This is an indication of the accuracy with which the disk result tracks the efficiency factor.

Table VI is a summary of data on EUREKA detector efficiency parameters deduced from the disk results. The quantity EFF is the average efficiency parameter deduced from 5 months (centered in mid-May 1983) of disk data acquired since early March 1983 when the detector positions in the shields became fixed. The standard deviation (1σ) in the (solid) data points plotted in Fig. 15 during this period is also given. The percent deviation between EFF and the original efficiency parameter (EFF_0) derived during the direct calibration at Los Alamos is tabulated for each detector. This latter quantity is a measure of the relative magnitude of the drift in the relative detection efficiency between the time the EFF_0 parameters were measured and mid-May 1983, approximately 6 months. Except for detector 4, for which this shift was anomalously large and negative (-7.5%), due to mispositioning the ^{241}Am source after reassembling the detector in the shield, the shift for the 6-month period varies between 1 and 5%. However, a 5% bias in the calculated count rate propagates to 10% in the concentration assay for uranium concentrations of 100 g/L (refer to Table V) and to 20% at 250 g/L. The gradual shifts are monotonic and positive for all detectors. Therefore, the corrections applied to the detector efficiency parameters have been essential in avoiding a large and increasing negative bias in the inventory results as a function of time.

The origin of the apparent decrease in the relative (186- to 60-keV) photopeak detection efficiencies is not defined. Since the effect is smooth and gradual with time, future efforts will incorporate the empirical trend for each detector into the EUREKA code. These will be used to automatically correct the efficiency parameters in future inventories. The disk measurements, performed at a greatly reduced frequency, will then be applied for measurement control purposes to compare with the empirically based predictions.

III. RESULTS OF SOLVENT EXTRACTION SYSTEM INVENTORY MEASUREMENTS

A. Introduction
Evaluation of the EUREKA inventory measurement capability included performing the pulsed and static EUREKA assay sequences on the solvent extraction system as described previously, followed by dumping of the solution components of

DETECTOR ID	EFF	1σ	$\dfrac{\left(EFF - EFF_0\right) \cdot 100}{EFF_0}$
1	2.460	1.3%	4.7%
2	1.750	0.8%	3.7%
3	2.275	0.5%	1.2%
4	1.886	0.4%	-7.5%
5	2.338	0.2%	2 5%
6	2.288	0.6%	3.8%

the system including external plumbing. The dumped components were analyzed externally for direct determination of solution volumes and masses of uranium. These direct results are the reference values used for comparison with the EUREKA inventory results.

EUREKA inventories were performed in 1983 during the first weeks of the months of February, March, April, May, August, and September. The corresponding direct measurements occurred with the February, March, May, and September inventories. The results are presented for February, March, April, and May. These results are given on a column-by-column basis for the extraction, strip, and backwash columns and for the plumbing associated with each column.

B. Direct Inventory Results
Following the EUREKA static measurements of uranium concentration in the separated phases in the isolated solvent extraction columns, the solution components of the solvent extraction system were dumped and analyzed. The column dumps included solution volumes in the pulser line through April 1983. Starting in May 1983, the pulser volumes were dumped and analyzed as separate components of the external plumbing. The volumes of the columns were drained (from the bottom) into six or seven 10-L bottles, and each bottle was weighed, homogenized, and sampled. The external plumbing components were handled similarly. The samples were analyzed for specific gravity (by a commercial, ultrasonic device) and for uranium concentration (by nondestructive, high-resolution gamma-ray assay[2,5]). Total volumes, determined from the measured weights and specific gravities, were used, along with uranium concentrations, to determine uranium masses. The estimated random relative uncertainty in the uranium concentration results is 0.7%, 1σ. The uncertainty in volume is approximately 1%, due to weighing. Therefore, the minimum overall random uncertainty in the direct inventory result for each column is approximately 1% for uranium mass and

approximately 0.5% for solution volume. The magnitudes of other possible random effects in the draining and sampling processes are unknown.

Prior to installation of EUREKA, a pilot effort to establish procedures for dumps, followed by external analysis of solvent extraction system solution components, was carried out (during July 1982) at Y-12. Following draining of the isolated solvent extraction columns (including the pulser lines), these columns were filled with a 50% HNO₃ solution. This flush solution was then drained and analyzed for uranium. Table VII summarizes the column inventory results of this pilot effort. The holdup, defined as the quantity of dissolvable uranium that remains in the column after the solution contents are dumped, is approximately 1.3% of the column inventory. The subsequent direct inventory measurements did not include such a flushing procedure, but it is assumed, for the purposes of comparison with EUREKA assays, that 1.3% must be added to the measured volumes and uranium masses of the dumped components.

Tables VIII and IX give the directly measured solution volumes and uranium masses (respectively) for the three solvent extraction system dumps. The quantities in parentheses incorporate the adjustments for holdup of solution in the columns following the dumps.

C. EUREKA Inventory Results

Figures 16 and 17 are plots of the uranium concentrations versus column height (where zero corresponds to the bottom of the column) measured during the pulsed and static EUREKA assays. The dashed and solid (plus dotted) lines are smooth curves drawn between the pulsed and static results, respectively. The solid histogram illustrates the method used by the EUREKA code to numerically integrate the results to give the column inventory. The solid vertical lines at the right of the histogram mark the heights of the aqueous-organic interface and the solution levels in the columns. These heights vary from column to column and month to month. The vertical position of detector 5 (5th detector from the bottom) differs for the pulsed and static assays for most inventory periods. Two (alternative) brackets are mounted on each column for this detector. The lower bracket, at the top of each stainless steel section, is used for all pulsed assays. In the static mode, because of the sharp discontinuity in concentration at the well-defined solution interface, it is desirable to position detector 5 below and as close to this interface as possible. Therefore, a second bracket for detector 5 is mounted for this purpose when the interface level is relatively high in the glass section.

TABLE VII

URANIUM INVENTORY HELD UP IN SOLVENT EXTRACTION COLUMNS FOLLOWING COLUMN DUMPS

COLUMN ID	COLUMN INVENTORY (G U)	COLUMN HOLDUP (G U)	HOLDUP · 100 / INVENTORY
EXTR.	5655	73.4	1.30%
STRIP	8778	120.5	1.37%
BACKW.	3132	39.2	1.25%
TOTAL	17565	233.1	1.33%

TABLE VIII

DUMP VOLUMES (ml)

DATE	COLUMN ID	COMPONENT Column	Pulser	Plumbing	Total
2/4/83*	EXTR	63845(64672)		19807(20064)	83650(84736)
	STRIP	62835(63651)		13549(13725)	76384(77376)
	BACKW	62624(63438)		8798(8396)	70922(71835)
	TOTAL	189302(191781)		41644(42185)	230946(233946)
3/4/83**	EXTR	62455(63765)		20279(20543)	82732(81800)
	STRIP	67505(68316)		13415(13589)	79918(78903)
	BACKW	62706(62508)		8516(8627)	71222(71235)
	TOTAL	186667(190089)		42210(42759)	228072(231844)
5/5/83	EXTR	60042(60825)	5350(5394)	20728(20997)	86120(85214)
	STRIP	59582(60357)	2619(2655)	13675(13800)	75826(76810)
	BACKW	58326(59652)	2802(2838)	8827(8962)	70555(71472)
	TOTAL	178550(180872)	10771(10885)	43178(43759)	230499(233498)

*COLUMN AND PULSER CONTENTS DUMPED SIMULTANEOUSLY

TABLE IX

DUMP INVENTORY RESULTS (GRAMS U)

DATE	COLUMN ID	COMPONENT Column	Pulser	Plumbing	Total
2/4/83*	EXTR	9318(9439)		2175(2201)	11493(11642)
	STRIP	9099(9217)		1506(1526)	10605(10745)
	BACKW	4314(4370)		752(782)	5066(5152)
	TOTAL	22731(23026)		4433(4491)	27164(27517)
3/4/83*	EXTR	5862(5918)		543(563)	6405(6503)
	STRIP	8359(8447)		1256(1272)	9595(9719)
	BACKW	3473(3518)		638(707)	4211(4225)
	TOTAL	17674(17905)		2497(2542)	21171(21445)
5/5/83	EXTR	3288(3331)	131(133)	1605(1626)	4908(4970)
	STRIP	9041(9159)	455(461)	1469(1488)	10965(11108)
	BACKW	3938(3985)	283(285)	827(838)	5048(5112)
	TOTAL	16267(16479)	749(759)	3901(3952)	20913(21190)

*COLUMN AND PULSER CONTENTS DUMPED SIMULTANEOUSLY

Large differences in the column concentration profiles for the pulsed and static measurements, particularly for the extraction column, illustrate the need for using pulsed assays to obtain concentration information that relates to the plumbing contents. The detector 1 (bottom) and detector 6 (top) assay results are used
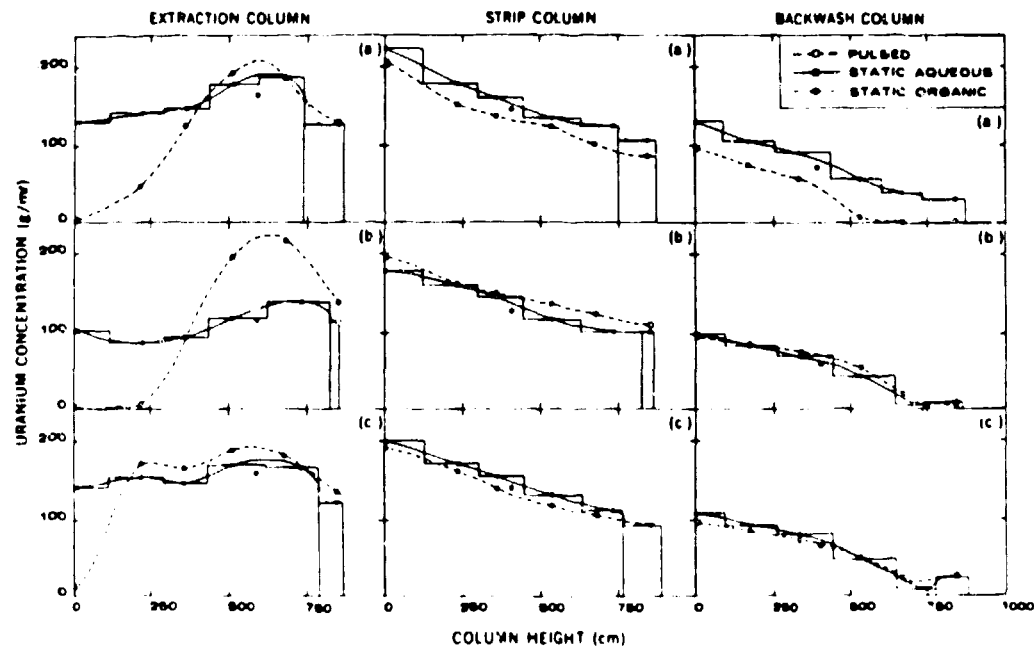
Fig. 16. Uranium concentration plotted versus column height for pulsed and static EUREKA assays at inventory. Smooth curves are drawn to approximate the pulsed and static profiles. The histogram used by the EUREKA code for inventory determination is also shown. Asterisks are external assay results for samples withdrawn from the static aqueous phase at a fixed position. The results correspond to the 1983 inventory measurements for (a) February, (b) March, and (c) April.

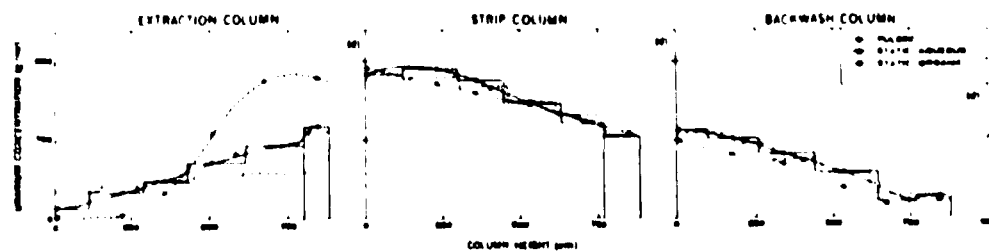Fig. 17. Uranium concentration plotted versus column height for pulsed and static EUREKA assays for the May 1983 inventory measurements. (Refer to Fig. 16 caption.) The dot-dash histograms are the result of the direct measurements of the dumped components of the column volumes. Since the pulser lines were dumped separately during this inventory, the comparison with the EUREKA histogram can be made.

for these purposes. Detector 1 is positioned below the organic feed line in the inactive region of each column. Therefore, it assays a pure aqueous solution during pulsed operation, and the concentration is representative of the aqueous solution drain components of the external plumbing for a given column. Detector 6 is mounted above the aqueous feed line in the inactive region of each column. Therefore, it assays a pure organic solution during pulsed operation, and the concentration is representative of the organic solution overflow components of the external plumbing for a given column.

For the pulsed EUREKA assays, no operator input is required other than column identification. For the static EUREKA assays, additional input is requested of the operator. This includes levels A and B of the solution and interface, respectively, in each column. These levels are read and recorded routinely by the operator at each inventory period after system shutdown and pulsing. The EUREKA code applies these quantities to the calculation of organic and aqueous solution volumes. The A level varies from month to month because samples are routinely pulled from the aqueous and organic phases following pulsing for phase disengagement. Since the EUREKA inventory is compared to the dump inventory, the A level after sampling is the value required by the code to compute the solution volume. The B level will also change following the sampling, but bigger factors in this change from month to month are the column operation parameters. Furthermore, should any shutdown (followed by pulsing) result in a B (interface) level below the glass sections, acid solution (free of uranium) is added to the column to raise this level to make it visible within the glass section. In all cases, the reading of A and B as well as the static EUREKA assays must follow the acid addition and sample withdrawal processes.

Because the operator routinely samples (for assay) the aqueous phase after shutdown and pulsing for phase disengagement, the EUREKA code is designed to use this assay information (uranium concentration and specific gravity), if it is available, to compute the acid molarity of the aqueous solutions in the column. This quantity had previously been observed to vary by as much as ± 2 M about a mean of 3.5 M in the extraction column. In this range (1.5 $\overline{M}$ to 5.5 M), the change in 186-keV count rate for 100 $\overline{g}$ U/L is approximately 2%. This propagates to a 4% effect in the uranium concentration assay. All EUREKA inventory results to date have employed the sample assay result in determining the correction for acid molarity in the extraction column. In the absence of this input to

the EUREKA code, the default values used by the code give rise to errors that are documented in Table II. Since the samples are withdrawn from the aqueous solutions at a fixed height in the column, the sample assay results are plotted at this height in Figs. 16 and 17.

Tables X and XI give the EUREKA results for solution volumes and uranium masses, respectively, for the inventories that included direct measurements. The quantities in parentheses adjacent to the column components incorporate corrections that have been applied recently to the original assay results. The corrections arise from the use (through May 1983) of an inaccurate quantity for the volume calibration (volume per unit column height) in the active regions of the columns. The correction amounts to approximately 2.5% and 2% on the column and system components, respectively, of solution volume and uranium mass.

Table XII gives the ratios of the results for the EUREKA volumes to those for the analogous dump components. (These are the ratios of the results in Table X to those in Table VIII.) The means and standard deviations in these quantities are given in Table XIII for the corrected ratios (numbers in parentheses in Table XII). Table XIV gives the ratios of the EUREKA results for uranium inventory to those for the analogous dump components. (These are the results of Table XI to those of Table IX.) The means and standard deviations in these quantities are given in Table XV for the corrected ratios (numbers in parentheses in Table XIV).

IV. RESULTS OF EVAPORATOR MONITOR EVALUATION

The uranium concentration monitor for the Y-12 secondary intermediate evaporator has functioned since installation as a device for readout of concentration; the monitor is used by the operator responsible for manual control of the evaporator. During several periods of operation of the monitor, samples were withdrawn from the return loop of the evaporator. The results of external analysis of the uranium concentration of these samples by nondestructive high-resolution gamma-ray assay[2,5] are plotted in Figs. 18 through 20 (as solid points) along with the monitor results (circles) at the assay cycle intervals. The percentage deviation between the monitor result (closest in time to the time of sample withdrawal) and the sample assay is plotted versus sample assay result in Fig. 21. These data indicate an average bias in the monitor result of approximately 1% with a relative standard deviation of 2% in the mean.

TABLE X

EUREKA VOLUMES (ML)

DATE	COLUMN ID	COMPONENT COLUMN*	PULSER**	PLUMBING**	TOTAL
2/4/83	EXTR	61206(59633)	2555	20423	84184(82611)
	STRIP	61497(59924)	2555	15772	79824(78251)
	BACKW	61933(60360)	2555	8691	73179(71606)
	TOTAL	184636(179917)	7665	44886	237187(232468)
3/4/83	EXTR	57749(58176)	2555	20423	82727(81154)
	STRIP	60964(59591)	2555	15772	79291(77718)
	BACKW	60867(59294)	2555	8691	72113(70540)
	TOTAL	181580(176861)	7665	44886	234131(229412)
5/5/83	EXTR	62418 (60845)	2555	20423	85396 (83823)
	STRIP	62224 (60651)	2555	15772	80551 (78978)
	BACKW	62272 (60699)	2555	8691	73518 (71945)
	TOTAL	186914 (182195)	7665	44886	259465 (234746)

*BASED ON CALCULATED COLUMN VOLUMES (USING DATA FROM ENGINEERING DRAWINGS)
AND ON OPERATOR INPUT OF HEIGHT OF SOLUTION AFTER SAMPLE WITHDRAWAL

**BASED ON CALCULATED VOLUME (USING MEASURED PIPE LENGTHS AND DIAMETERS)

TABLE XI

EUREKA INVENTORY RESULTS (GRAMS U)

DATE	COLUMN ID	COMPONENT COLUMN	PULSER	PLUMBING	TOTAL
2/4/83	EXTR	9255(9017)	10	1836	11101(10863)
	STRIP	9396(9156)	526	1648	11570(11330)
	BACKW	4452(4339)	242	569	5263(5150)
	TOTAL	23103(22512)	778	4053	27934(27343)
3/4/83	EXTR	6673(6497)	4	1947	8624(8448)
	STRIP	8166(7955)	501	1946	10613(10407)
	BACKW	3231(3148)	247	600	4078(3995)
	TOTAL	18070(17600)	752	4493	23315(22845)
5/5/83	EXTR	3946(3847)	10	2486	6442(6343)
	STRIP	9600(9357)	489	1924	12013(11770)
	BACKW	4144(4039)	251	646	5041(4936)
	TOTAL	17690(17243)	750	5056	23496(23049)

TABLE XII

(EUREKA VOLUME)/(DUMP VOLUME)

DATE	COLUMN ID	COMPONENT COLUMN	PULSER	PLUMBING	TOTAL
2/4/83*	EXTR	0.999(0.962)		1.031(1.018)	1.006(0.975)
	STRIP	1.019(0.982)		1.164(1.149)	1.045(1.011)
	BACKW	1.010(0.992)		1.049(1.035)	1.032(0.997)
	TOTAL	1.016(0.978)		1.078(1.064)	1.027(0.994)
3/4/83*	EXTR	0.998(0.960)		1.007(0.994)	1.000(0.968)
	STRIP	1.016(0.978)		1.176(1.161)	1.044(1.011)
	BACKW	1.028(0.989)		1.021(1.007)	1.027(0.992)
	TOTAL	1.014(0.976)		1.063(1.050)	1.023(0.985)
5/5/83**	EXTR	1.025(0.987)		0.936 (0.973)	1.015 (0.984)
	STRIP	1.041(1.003)		1.158 (1.143)	1.062 (1.028)
	BACKW	1.050(1.012)		0.985 (0.972)	1.042 (1.002)
	TOTAL	1.039(1.002)		1.040 (1.026)	1.039 (1.005)
5/5/83	EXTR	1.042(1.000)	0.763(0.753)		
	STRIP	1.044(1.002)	0.976(0.963)		
	BACKW	1.057(1.017)	0.912(0.900)		
	TOTAL	1.047(1.007)	0.874(0.863)		

*COLUMN AND PULSER CONTENTS DUMPED SIMULTANEOUSLY

**SEPARATED COLUMN AND PULSER COMPONENTS COMBINED FOR THIS ANALYSIS

TABLE XIII

AVERAGE VALUES (±1σ) OF (EUREKA VOLUME)/(DUMP VOLUME)

COLUMN ID (NO AVERAGED)	COLUMN + PULSER	PLUMBING	TOTAL
EXTR (3)	0.970 ± 0.015	0.995 ± 0.073	0.972 ± 0.008
STRIP (3)	0.988 ± 0.013	1.159 ± 0.012	1.017 ± 0.010
BACKW (3)	0.998 ± 0.013	1.005 ± 0.037	0.999 ± 0.008
ALL (9)	0.985 ± 0.017	1.050 ± 0.078	0.997 ± 0.019

V. DISCUSSION AND RECOMMENDATIONS

A. EUREKA

1. Column Inventory Results. Exercise of a rigorous measurement control program during the first six months of operational evaluation of EUREKA has generated confidence in the ability to establish and maintain an accurate calibration for the concentration assay. However, comparison of the EUREKA inventory results with direct measurements indicates an average bias of approximately 3% in the EUREKA column inventories (Table XV). The standard deviation in the relative results is 6%. The bias is large compared to the relatively good agreement in column solution volumes (-1.5% on the average).

The largest inventory bias is observed for the extraction column--in particular, for the

May 1983 measurements for which the EUREKA result was 15% larger than the direct result. Elimination of the May 1983 measurement from the average results in a bias of 1.8% and a standard deviation in the relative result of 4.5%. The distinctly flatter uranium concentration profile deduced from the dumped component assays compared to the EUREKA assays (dot-dash compared to solid histograms, Fig. 17) suggests that changes (resulting, perhaps, from equilibration of concentration) occurred between the EUREKA measurements of the static extraction column and the actual dump of the extraction column during the May 1983 inventory. However, no changes that might account for the apparent bias have been documented.

TABLE XIV

(EUREKA INVENTORY)/(DUMP INVENTORY)

DATE	COLUMN ID	COLUMN	PULSER	PLUMBING	TOTAL
2/4/83*	EXTR	0.994(0.956)		0 844(0.833)	0.966(0.933)
	STRIP	1.090(1.050)		1.094(1.080)	1.091(1.055)
	BACKW	1.088(1.048)		0.757(0.747)	1.033(1.004)
	TOTAL	1.051(1.011)		0.914(0.902)	1.028(0.994)
3/4/83*	EXTR	1.139(1 095)		1.262(1.246)	1.165(1.126)
	STRIP	1.039(1 001)		1 549(1 530)	1.106(1 070)
	BACKW	1.001(0.565)		0.860(0.849)	0.978(0.946)
	TOTAL	1 065(1 025)		1.285(1 268)	1 101(1.065)
5/5/83**	EXTR	1 198(1 154)		1 549(1 529)	1 313(1.276)
	STRIP	1 062(1 023)		1 310(1 293)	1 096(1.060)
	BACKW	1.042(1.004)		0.781(0.771)	0.999(0.966)
	TOTAL	1 084(1 044)		1 296(1 279)	1 125(1.088)
5/5/83	EXTR	1 200(1 155)	0 769(0.769)		
	STRIP	1 062(1 022)	1 075(1.061)		
	BACKW	1.052(1.013)	0 893(0.881)		
	TOTAL	1 087(1 046)	0 999(0 988)		

*COLUMN AND PULSER CONTENTS DUMPED SIMULTANEOUSLY

**SEPARATED COLUMN AND PULSER COMPONENTS COMBINED FOR THIS ANALYSIS

TABLE XV

AVERAGE VALUES (±1σ) OF (EUREKA INVENTORY)/(DUMP INVENTORY)

COLUMN ID (NO. AVERAGED)	COLUMN + PULSER	PLUMBING	TOTAL
EXTR (3)	1 068 ± 0 101	1 205 ± 0 350	1 111 ± 0 172
STRIP (3)	1 025 ± 0 025	1 301 ± 0 225	1 062 ± 0 008
BACKW (3)	1 006 ± 0 042	0 789 ± 0 051	0 972 ± 0 029
ALL (9)	1 032 ± 0 065	1 098 ± 0 315	1 048 ± 0 107

The measured standard deviation of 6% is twice as large as the predicted random uncertainty in the EUREKA inventory of a given column. However, 6% is the standard deviation in the relative result, and therefor it includes random effects associated with dumping the columns, sampling the dumped components, and performing external assays of the components. Similarly, the 3% apparent bias is affected by the systematic effects in these processes of direct verification. For future comparisons, efforts will be directed at reducing the magnitude of possible random and systematic effects from the direct verification procedures. This includes assuring that liquid levels are recorded after (rather than before) sampling lines are sparged and samples are withdrawn, that all valves to the external plumbing are closed and remain closed after shutdown, and that no additional solution is added to or removed from the columns between the EUREKA assays and the dumps. (The addition of acid or de-ionized water to the columns to raise the interface level in the column is a routine practice during inventory.)
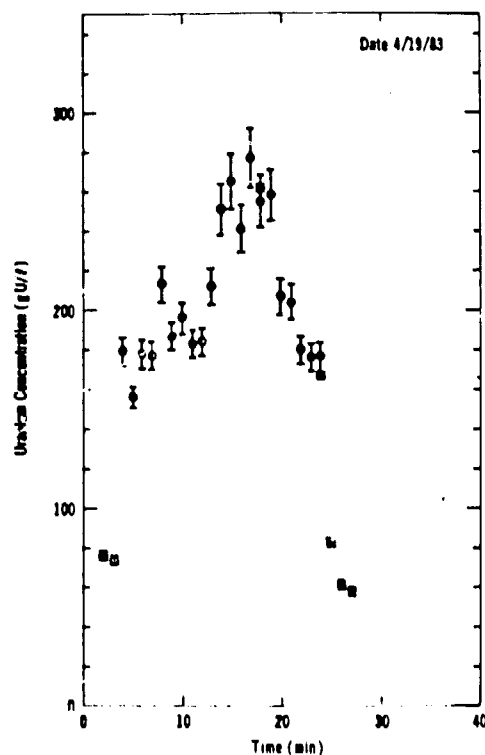

Fig. 18. Evaporator monitor results for uranium concentration plotted versus time. The results of the external analysis of two samples withdrawn from the evaporator return loop are also plotted at the time of sample withdrawal (solid points). The error bars are calculated from the counting statistics (1σ) of the monitor assays.

2. Concentration Assays. The asterisks plotted in Figs. 16 and 17 are the concentrations of uranium samples withdrawn from the static columns at the indicated height and assayed externally. In all cases, these samples were withdrawn from the columns before the static measurements were performed but after the pulsing of the isolated columns for phase separation was completed. The asterisks fall below the solid smooth curve that connects the static assay data in Fig. 16. The average discrepancy between the smooth curve and the sample results is 10% (±4%, 1σ).

Several possible causes for these discrepancies have been investigated. Among these are
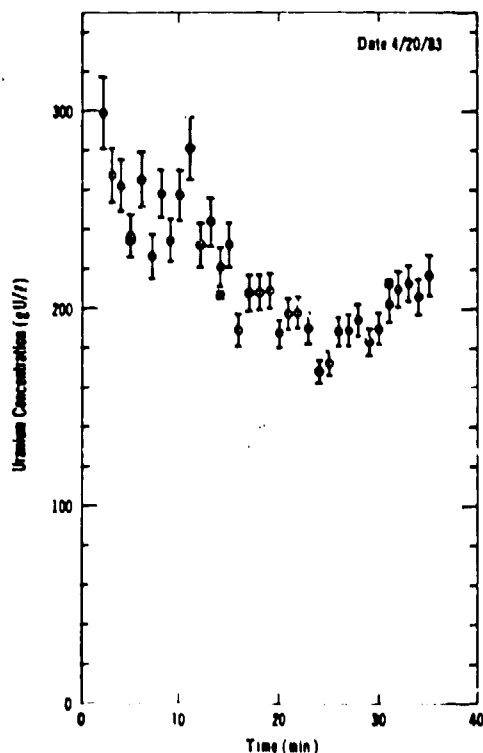
Fig. 19. Evaporator monitor results for uranium concentration plotted versus time. The results of the external analysis of three samples withdrawn from the evaporator return loop are also plotted at the time of sample withdrawal (solid points). The error bars are calculated from the counting statistics (1σ) of the monitor assays.
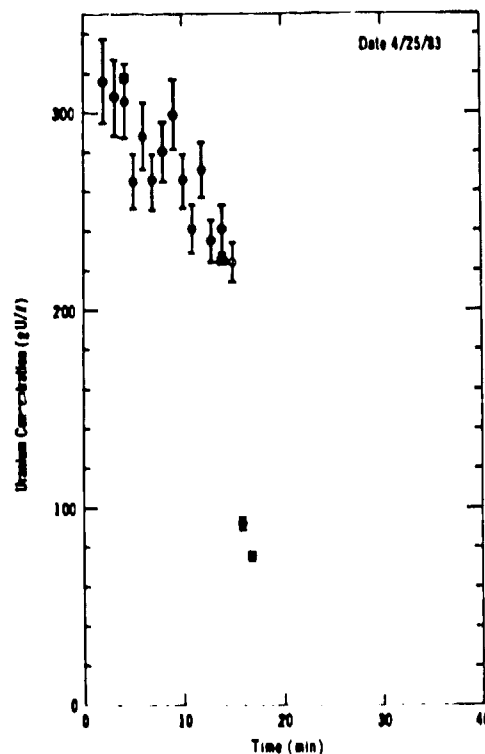

Fig. 20. Evaporator monitor results for uranium concentration plotted versus time. The results of the external analysis of two samples withdrawn from the evaporator return loop are also plotted at the time of sample withdrawal (solid points). The error bars are calculated from the counting statistics (1σ) of the monitor assays.

the calibrations of the concentration assay for each detector. The standard cell was moved out to the operations area in order to perform the direct calibration of each detector at its normal location in the solvent extraction system. The calibration results (open circles in Fig. 15) show that the foil assays track the detector efficiencies to within 0.5%, dismissing calibration of the concentration assay as the cause of the discrepancies.

Contamination on the outer surfaces of the columns was also dismissed as a possible cause by performing the EUREKA assays (on the static columns), decontaminating the outer surfaces of the columns, and running the assays again. Contamination of the inner surfaces of the columns

with insoluble HEU was another possibility, but this was dismissed when the May 1983 inventory results (Fig. 17) showed agreement between the smooth curve drawn between the static EUREKA assay results and the results of the aqueous sample assays (asterisks).

To date, the cause of the discrepancies has not been identified. The average magnitude of the difference is substantially larger than the average bias (3%) in the EUREKA column inventory results. This suggests that the observed discrepancy is probably not caused by systematic effects that bias the EUREKA assay, but rather by effects of sampling the column or by incorrect assumptions about the sampling (for example, plotting the sample assay results at the height of the sample spigot).
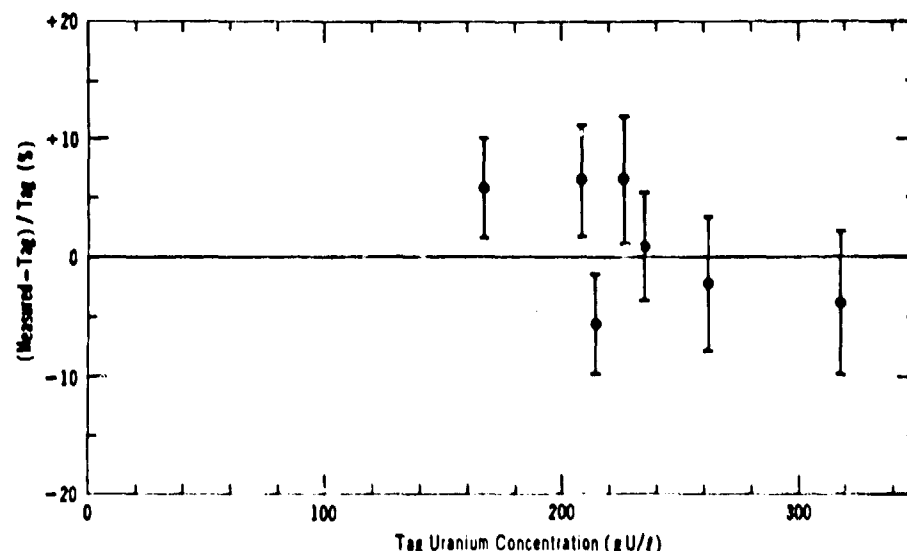
Fig. 21. Percentage deviation between evaporator monitor result ("meas-
urement") and external sample assay result ("tag") plotted versus exter-
nal sample assay result for the seven withdrawn samples indicated in
Figs. 18 through 20. The error bars are calculated from the counting
statistics (1σ) of the single monitor assay concurrent with the sample
withdrawal.

Because uranium is partitioned between two
immiscible phases in the solvent extraction
columns, a gradient in concentration (versus
column height) will always exist within a phase,
even after long periods of pulsing following
shutdown and isolation of the columns. This
gradient contributes to the difficulty of using
sampling methods to evaluate the EUREKA concen-
tration assay. For the same reason, determina-
tion of HEU inventory in the solvent extraction
columns by sampling is subject to large uncer-
tainties. The choice of the on-line NDA methods
applied in the EUREKA instrument was made, in
part, to overcome the uncertainties in the in-
ventory results introduced by sampling.

The concentration monitor on the Y-12 in-
termediate evaporator return loop may offer a
more reasonable situation for evaluation of the
concentration assay by sampling since only a
single-phase solution is present in the evapo-
rator.

3. Inventories Obtained During Pulsed
Operation. The EUREKA instrument performs rou-
tine assays of uranium concentration during
pulsed operation, primarily to deduce the ura-
nium concentrations (and hence the HEU inven-

tory) in the plumbing external to the secondary
solvent extraction system. Because the uranium
concentration profile is measured during pulsed
operation by the EUREKA detectors, the results
of these assays can also be used to deduce the
HEU inventory in the operating columns. Fur-
thermore, if steady-state operation of the col-
umns is achieved at the time of the EUREKA as-
says during pulsed operation, and steady state
persists until the time of shutdown, then the
EUREKA pulsed and static inventories should be
the same.

The use of the assay data obtained during
pulsed operation for determination of the HEU
inventory in the solvent extraction columns is
appealing because it opens the possibility of
using a mathematical model[6] to give an accu-
rate concentration profile while using substan-
tially fewer than six detectors per column. The
use of such a model, however, requires that
steady-state operation persists from the time
of the measurement of concentrations until the
time of shutdown.

Table XVI is a tabulation of the HEU in-
ventories in the three columns determined from
the pulsed and static EUREKA assays for the four

TABLE XVI

EUREKA PULSED AND STATIC COLUMN INVENTORIES (GRAMS U)

DATE	COLUMN ID	PULSED	STATIC	STATIC/PULSED	ΔT(HOURS)
2/4/83	LX1R	7214	9255	1.28	20
	STRIP	8127	9396	1.16	20
	BACKW	2205	4452	2.02	20
	TOTAL	17546	23103	1.32	
3/4/83	LX1R	6748	6673	0.99	1.5
	STRIP	8897	8166	0.92	0.5
	BACKW	3462	3231	0.93	0.25
	TOTAL	19107	18070	0.95	
4/8/83	LX1R	9014	9346	1.04	5
	STRIP	8276	8849	1.07	5
	BACKW	3610	3747	1.04	5
	TOTAL	20900	21942	1.05	
5/5/83	LX1R	5488	3946	0.72	3
	STRIP	9336	9600	1.03	3
	BACKW	3236	4144	1.28	3
	TOTAL	18060	17690	0.98	

inventory periods. The quantity ΔT is the time elapsed between the pulsed assay and the actual column shutdown. The largest differences between static and pulsed inventories appear in the February results, which correspond to the longest time delay between pulsed assays and shutdown, but substantial differences also exist in the May results for which the delay was only 3 h.

Production routines are such that there can be no guarantee of steady-state operation of the solvent extraction systems. Furthermore, the routine procedure of adding acid (or de-ionized water) to the columns after shutdown and phase separation in order to raise the interface to a height above the glass-stainless steel flange alters the column HEU inventory from that at the time of shutdown. Therefore, the ability to use the concentration assays in the pulsed mode (and hence the usefulness of a mathematical model in simplifying the EUREKA inventory determination) cannot be evaluated using the results obtained in these exercises. For the present, a significant reduction in the number of concentration measurements (that is, the number of detectors) required to obtain the HEU inventory in a given column is not possible.

4. System Inventory Results. The HEU inventories deduced by EUREKA for the plumbing external to the solvent extraction columns are larger than the dump results by 10%, on the average (refer to Table XV). Since the plumbing inventory is only approximately 20% of the total

inventory, the additional bias introduced in the total EUREKA inventory is only 2%.

Some of the systematic bias must be attributed to changes in concentrations in feed and product streams that occur between the time of the pulsed EUREKA assays and the shutdown; these are due to lack of steady-state operating conditions. At least one half of the average bias (that is, 5%) is the result of an overestimate by EUREKA of the solution volumes in the external plumbing. (Refer to Table XIII.) The volumes assumed by EUREKA are obtained by calculations using the known diameters and lengths of pipe in the external plumbing. It is assumed that these plumbing volumes are full of solution at inventory, but for some of the external components, this appears to be a false assumption.

To date the bias in the EUREKA solvent extraction system inventory is 5% with a standard deviation of 10% in the relative results. Improvements can be expected by using more realistic volumes for the external plumbing components.

5. Recommendations. The following procedures are recommended for the remaining period of operational evaluation of the EUREKA instrument:

• Increase efforts to reduce uncertainties in direct inventory verification methods. This includes valving off external plumbing lines immediately after shutdown of solvent extraction system, sparging sample lines and withdrawing samples before reading liquid levels in columns, performing static EUREKA assays as soon possible after samples are withdrawn, and uring that addition (or removal) of any solution to (or from) the columns precedes the EUREKA assays of the static columns.

• Use the empirical results of the column dumps to reevaluate the volumes of the individual external plumbing components. This will reduce the EUREKA bias that results from the assumption that the external plumbing volumes are full of solution at inventory.

• Reduce the time between EUREKA assays of the pulsed columns and the actual shutdown. This will increase the accuracy of the EUREKA determination of the HEU inventory in the external plumbing.

The following changes are recommended for improving the effectiveness of the EUREKA instrument as a tool in a production facility:

- Reduce measurement control efforts substantially. The extensive measurement control results accumulated to date suggest that the background results and the uranium disk assay results are independent of column identification and operational mode (static or pulsed). Since the detector efficiencies change slowly with time, a disk assay is required before each inventory. However, a disk assay is not necessary before each column assay. It is recommended that the number of disk assays be reduced from six to one per detector for each inventory period, and that the background measurements be performed only once (rather than twice) on each column.

- Eliminate the withdrawal and external assay of samples of t e aqueous and organic phases of the contents of the solvent extraction columns at inventory.

- Reduce the number of detectors required per column from six to five or fewer.

- Modify EUREKA hardware and software to permit simultaneous assay of the three columns of the secondary solvent extraction system.

- Implement the other secondary solvent extraction system with hardware to enable the use of EUREKA for determining the HEU inventory in this parallel system.

B. Evaporator Monitor

The uranium concentration monitor for the Y-12 secondary intermediate evaporator has performed accurately and reliably since its installation in December 1982. Because these performance criteria have been satisfied, the possibility of automatic control of the evaporator operation can now be considered.

The desired product solution for feed into the secondary solvent extraction system is well defined in uranium concentration as well as acid molarity. For this reason, combined on-line measurements of uranium concentration and specific gravity might be used as a basis for automating control of the evaporator to achieve the optimal product.

Some progress has been made at Y-12 in evaluation of on-line specific gravity measurements and automatic control of evaporator oper-

ation using on-line results.[a] One approach to obtaining automatic control for this evaporator is to apply the on-line results of these two demonstrated methods to the existing Y-12 automation mechanism.

It is recommended that the accuracy of the evaporator monitor be verified by withdrawal and external analysis of samples of solution in the return loop as long as the evaporator continues operation in the manual-control mode.

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[a]This information was furnished by J. Lanford, Oak Ridge Y-12 Plant, 1983.