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DEVELOPMENT OF IN-LINE PLUTONIUM SOLUTION NDA INSTRUMENTATION AT THE SAVANNAH RIVER PLANT

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Abstract

A K-absorption-edge plutonium solution densitometer has been developed by the Los Alamos Scientific Laboratory for in-line test and evaluation in the process line at the Savannah River Plant. The design features of the measurement system, the measurement principles, and performance data are presented.

1. Introduction

The K-edge Pu solution densitometer for in-line test and evaluation at the Savannah River Plant (SRP) has been fabricated and is undergoing final testing at LASL. The instrument is to be used in a cooperative test and evaluation exercise by LASL, SRP, and DOE and is scheduled for shipment to SRP during April, 1980. It will first be installed off-line for preliminary testing before receiving approval from SRP for full in-line installation. The eventual assay task involves the measurement of the total Pu concentration in the precipitator feed solution while it resides in the cation exchange holding tanks of the product line at SRP.

2. Measurement Principles

The assay instrument employs the technique of K-edge absorption densitometry, which measures the total concentration of an individual element in a sample of well-defined geometry. The Pu concentration assay by the SRP densitometer involves the use of radioactive sources of ^{75}Se and ^{57}Co which provide gamma rays of energies 121.1 and 122.1 keV, respectively. These gamma rays pass through a sample cell containing the Pu solution, facilitating the measurement of the differential transmission of photons across the 121.8-keV Pu K absorption edge. The fraction (T) of the incident photon intensities at 121.1 and 122.1 keV which are transmitted by the sample material are determined by comparison of these sample spectra with ^{75}Se and ^{57}Co spectra acquired previously during a regular measurement control run with an empty cell. From the photon transmission above the Pu K edge (T_U) and below the K edge (T_L), the Pu concentration in the sample solution is given by $k\ln(T_L/T_U)$, where k is a matrix-independent calibration constant determined by the photon absorption characteristics of Pu and the geometrical features of the sample cell.

The isotopic composition of the sample material can be inferred from passive gamma-ray counting of the solution through an enlarged collimator. The most useful gamma-ray energy range for this densitometer is limited to between 60 and 300 keV by detector efficiency and window thicknesses in the instrument. In this energy range it is possible to obtain relative weight per cent ratios for $^{238}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ by measurement of the ratios of the areas of the following gamma-ray peaks (after correction for sample self-absorption): 153 keV/129 keV and 148 keV/129 keV, respectively. The total solution isotopic composition is then determined by first postulating the ^{239}Pu weight per cent and using isotopic correlations to obtain the ^{240}Pu and the ^{242}Pu weight fractions. Then all of the isotopic weight per cents are computed (including a re-computation of the ^{239}Pu fraction), subject to the consistency of the result with the measured 238/239 and 241/239 ratios. This procedure is repeated until convergence is achieved. With the relatively narrow range of isotopics to be encountered at SRP, this method is expected to work quite well with very few iteration cycles.

3. Instrument Design

The radioactive sources are positioned on a wheel which shares a common axle with a second, collimator wheel and is driven by a motorized Geneva drive mechanism. Pu process solution is pumped from one of the holding tanks into a stainless steel transmission cell which is situated inside an extension of the process cabinet containment barrier. A schematic illustration of the complete in-line installation is shown in Figure 1, where the process holding tanks, solution

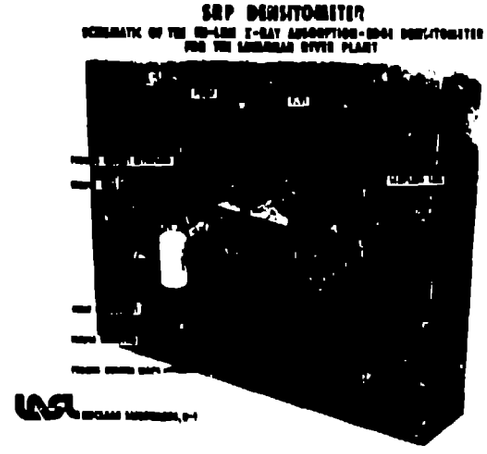

Fig. 1. Schematic of the in-line installation configuration for the Pu solution densitometer at the Savannah River Plant.

plumbing, and measurement station are indicated. The sample cell and its containment extension are located between the source and collimator wheels, in a standard transmission geometry. All components of the assay instrument except the sample cell are located outside the process cabinet containment for convenient accessibility and minimal contamination of the instrument. The use of the K-edge technique permits the use of sturdy containment barriers between the sample cell and the instrument, because of the emphasis on gamma radiation with adequate penetrability. A sketch of the measurement station is shown in Figure 2, where the position of the assembly on a modified panel of the process cabinet is depicted. The entire structure shown will be supported at a height of two meters from the floor, on the operations side of the process cabinet, thereby allowing adequate head room for operations personnel. Pulse-processing electronics, along with the data analysis and display equipment, will be located across from the process cabinet.

The source and collimator wheels are designed with a third position in which the radioactive sources are rotated behind shielding and the detector views the sample cell through an enlarged collimator. In this configuration, the instrument can acquire a spectrum of the gamma radiation emitted by the sample solution itself. From these data, the instrument sorts out the contributions to the spectrum from each Pu isotope and infers the isotopic composition of the sample by the method described above.

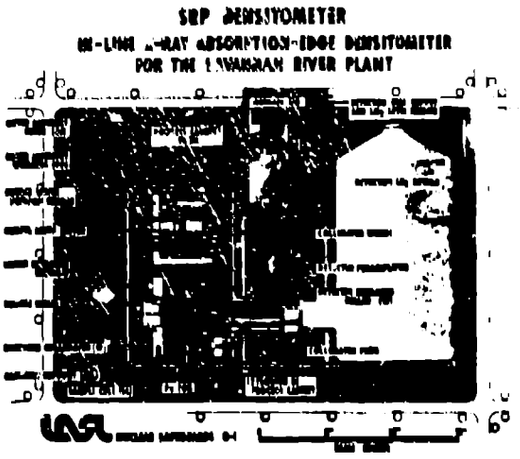

Fig. 2. Illustration of the measurement station for the in-line Pu solution densitometer at the Savannah River Plant. The mechanism for the movement of the source and collimator wheels (shown at the left) straddles the process cabinet extension so that the sample cell (inside the process cabinet containment) is between the transmission sources and the detector.

4. Software Design

All assay sequences are fully automated and are administered by an LSI-11 minicomputer, which is an integral part of a Nuclear Data ND/660 Multichannel Analyzer System. The computer-based nature of this NDA instrument facilitates both the automatic checking of the system for gain shifts, detector efficiency and resolution stability, and also the application of corrections to the raw data for source decays and rate-related losses in the electronics.

The densitometer is connected to the process through a computer-controlled bypass solution plumbing system which permits the transport of the solution to the assay cell from either of the holding tanks or from an outside source. Furthermore, provision is made for draining of the cell contents into an intermediate reservior after assay to allow the pulling of a sample of the assayed material for off-line chemical and/or mass-spectrometric analysis. The reservior contents are then routinely drained back to one of the holding tanks after computer verification that this last drain will not interrupt the flow of the process.

For every assay sequence, a specific set of valve settings in the plumbing system is expected by the computer. Eight of the ten valves in the plumbing circuit are solenoid valves and are operated by the computer in accordance with the assay task requested. There are also two manual valves which are set by the operator in choosing from where the sample material will be obtained. Provision is also made, through an electronic interface between the computer and the plumbing circuit, for manual operation of all ten valves, should the need arise. The computer reads the status of all ten valves through the electronic interface and checks the settings for correctness to ensure the proper transport of solution through the system without interference with the process operation. At each step in an assay sequence, if an incorrect valve setting is noted, the computer advises the operator of the fault and will not proceed until the correct valve configuration is achieved. A sample assay sequence is given below:

EXAMPLE ASSAY SEQUENCE

1. Operator types in password to unlock the keyboards, and the computer returns with a request for the assay option.
2. The operator types in an assay request. The request can be made in one typewritten line, giving all the necessary assay parameters, or in a "computer-question, operator-answer" dialog.
3. The computer types instructions to the operator concerning the required manual valve settings, based upon the operator's assay request.
4. The computer verifies the operator's valve settings and then sets the automatic valves for the assay sample material requested.

5. The pump is turned on, and a "solution flow" signal from a flowmeter indicates that the sample cell has filled.
6. Solution is circulated for a brief period to charge the plumbing lines and sample cell with fresh solution.
7. Solution flow is then stopped by closing all automatic valves and turning off the pump.
8. The assay of the static cell contents is carried out.
9. After completion of the assay, the operator is called back to the computer terminal, and the cell contents are drained to the intermediate reservior.
10. The operator is given the opportunity to pull a sample of the assayed material for off-line analysis.
11. After the sample is pulled, or if the sample is refused, the reservior is drained back to the process, and then all valves are closed.
12. The computer keyboard is locked and the machine is returned to an "idle" state, awaiting a password entry to initiate the procedure again.

In support of this assay sequence, certain measurement control procedures are available. First, periodic measurements are made of the unattenuated ^{75}Se and ^{57}Co spectra (i.e., transmitted through an empty cell). These empty-cell measurements are compared with previous ones to check the stability of the densitometer geometry and calibration. The computer reminds the operator of the need for a repeat of these empty-cell measurements when two or more days have elapsed since the last such run.

Verification of the unattenuated gamma-ray intensities is followed by a densitometry measurement of a standard Pu foil which is permanently installed in the densitometer. The foil can be pulled reproducibly into an assay position so that its effective Pu concentration can be measured and compared with a previously-determined value. The foil assay procedure serves as a convenient secondary calibration check between full-scale calibration runs. Failure of a given foil assay to compare favorably with the stored value will result in the flagging of all subsequent densitometry results as questionable because of failure of this measurement control test. This labeling of the results will continue until the foil test is repeated and passed successfully.

The densitometer software package has been written to require minimal operator interaction and to facilitate reliable operation of the instrument with negligible perturbation of the process schedule and maximum physical and radiation safety for plant and operations personnel. This is accomplished by programming the automatic application of a variety of diagnostic tests, the clear counseling of the operator if any of these tests is failed and action is required, and the execution of the assay if the outcome of all of these tests is positive. This extra measure of human engineering expended to accomplish these goals constitutes the major component of the design efforts for this in-line instrument.

5. Preliminary Test Results

Test assays at LASL of the standard Pu foil over extended periods of time have demonstrated the overall stability of the instrument. Foil assay test data are shown in Figure 3, where the 0.3% instrument precision shown was achieved in assay count times on the order of 30-40 minutes. In Figure 4 we show some densitometry results on test cells of Pu solution at LASL. Each data point represents an average of several runs on samples at the solution concentrations indicated, and thus reflect higher precision than routinely achieved in one measurement. For a single densitometry assay lasting 30-40 minutes, precisions on the order of 0.5% or better can be achieved.

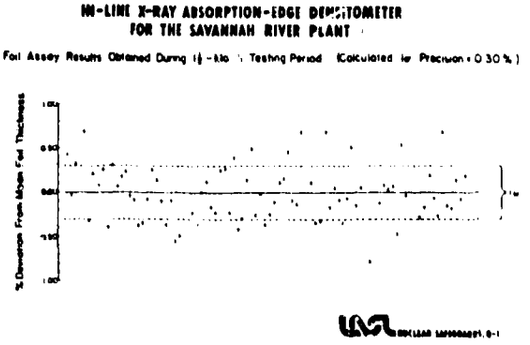

Fig 3. Fluctuation in effective Pu concentration measured for a (secondary) standard Pu foil installed in the densitometer.

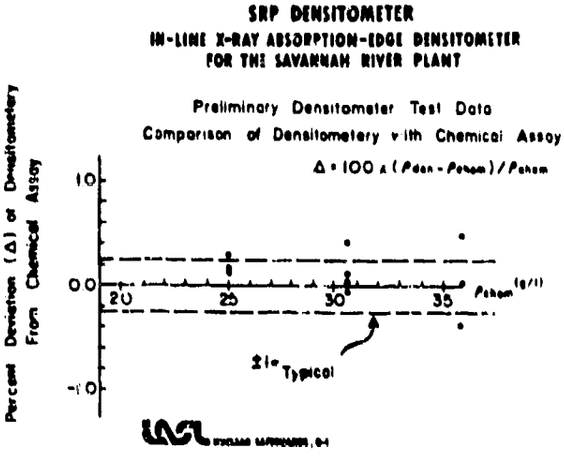

Fig. 4. Densitometry results on standard solutions during pre-shipment tests at LASL. The concentrations used in these test runs span the range of Pu concentrations to be encountered at SRP.

[It should be noted that, for a given set of counting times, better precision is obtained in a foil assay than in a solution assay. This is a result of the lower solution assay count rates caused by the added gamma-ray attenuation by the solution.]

Final isotopics calibration of the instrument must be performed on SRP solution samples to establish the proper isotopic correlations. However, preliminary isotopics calibration tests at LASL show approximately a 15 % precision on the measurement of the $^{238}Pu/^{239}Pu$ weight ratio and approximately 2 % precision on the $^{241}Pu/^{239}Pu$ ratio in 20-minute passive counts on weapons grade samples. These data indicate that we can expect to be able to specify the ^{240}Pu weight fraction in assay samples with approximately a 3 % precision in the same counting time.

6. Conclusion

Pre-shipment tests of the densitometer at LASL have demonstrated the overall soundness of the hardware designs, the stability of the assay results, and the thoroughness and reliability of the computer software. After shipment to SRP, the instrument will first be installed on a designated glove box in the SRP analytical laboratory area, where all possible operational modes of the system and its interaction with the process through the plumbing system will be tested and where operations personnel will receive preliminary training in its use. In-line installation is tentatively scheduled for late FY 1980, after completion of all off-line tests.