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TITLE: ROBOTIC SAMPLE PREPARATION FOR RADIOCHEMICAL PLUTONIUM AND AMERICIUM ANALYSES

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ROBOTIC SAMPLE PREPARATION FOR RADIOCHEMICAL PLUTONIUM AND AMERICIUM ANALYSES*

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ABSTRACT

A Zymate robotic system has been assembled and programmed to prepare samples for plutonium and americium analyses by radioactivity counting. The system performs two procedures: a simple dilution procedure and a TTA (xylene) extraction of plutonium. To perform the procedures, the robotic system executes 11 unit operations such as weighing, pipetting, mixing, etc. Approximately 150 programs, which require 64 kilobytes of memory, control the system. The system is now being tested with high-purity plutonium metal and plutonium oxide samples. Our studies indicate that the system can give results that agree within 5% at the 95% confidence level with determinations performed manually.

INTRODUCTION

The Analytical Chemistry Group at the Los Alamos National Laboratory provides analytical support to the plutonium recovery operation run by the Laboratory. The Group analyzes thousands of samples per year from process streams, intermediate products, and final plutonium products. Most of the analyses are done by established procedures. The routine nature of these analyses and a desire to learn firsthand about the capabilities of robotic systems in the analytical chemistry laboratory prompted an interest in using a robotic system to automate sample preparation procedures.

A Zymate robotic system has been assembled and programmed to prepare samples for plutonium and americium analyses by radioactivity counting. The system can perform two procedures: (1) a simple dilution procedure and (2) a TTA (xylene) extraction of plutonium away from the americium. Results to date indicate that the robotic system, which is now being tested with high-purity plutonium metal and plutonium oxide samples, can perform the preparation procedures with the desired accuracy and precision.

DESCRIPTION OF SAMPLE PREPARATION PROCEDURES

The preparation of samples for plutonium and/or americium analyses by radioactivity counting, i.e., the counting of alpha and gamma

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activity, is done by either a simple dilution procedure or an extraction-dilution procedure. A simplified flow diagram of the procedures is shown in Figure 1. Details of the extraction method can be found in Reference 1. The aliquots taken for gamma counting are placed in a test tube, and the aliquots taken for alpha counting are dried on a 1" square glass cover slip. The alpha activity is measured in gas flow proportional counters, and the americium gamma activity at 59.6 keV is measured in an automatic gamma counter with the window set at 35 to 80 keV. Knowing the gamma activity from the americium allows a correction to the gross alpha activity, which then gives the alpha activity caused by plutonium. If the americium alpha activity is $\geq$90% of the gross alpha activity, an extraction procedure is done to obtain a more accurate plutonium value. Regardless of the preparation procedure for plutonium, the americium is always measured by the dilution method.

FLOW DIAGRAM FOR DILUTION AND EXTRACTION PROCEDURES

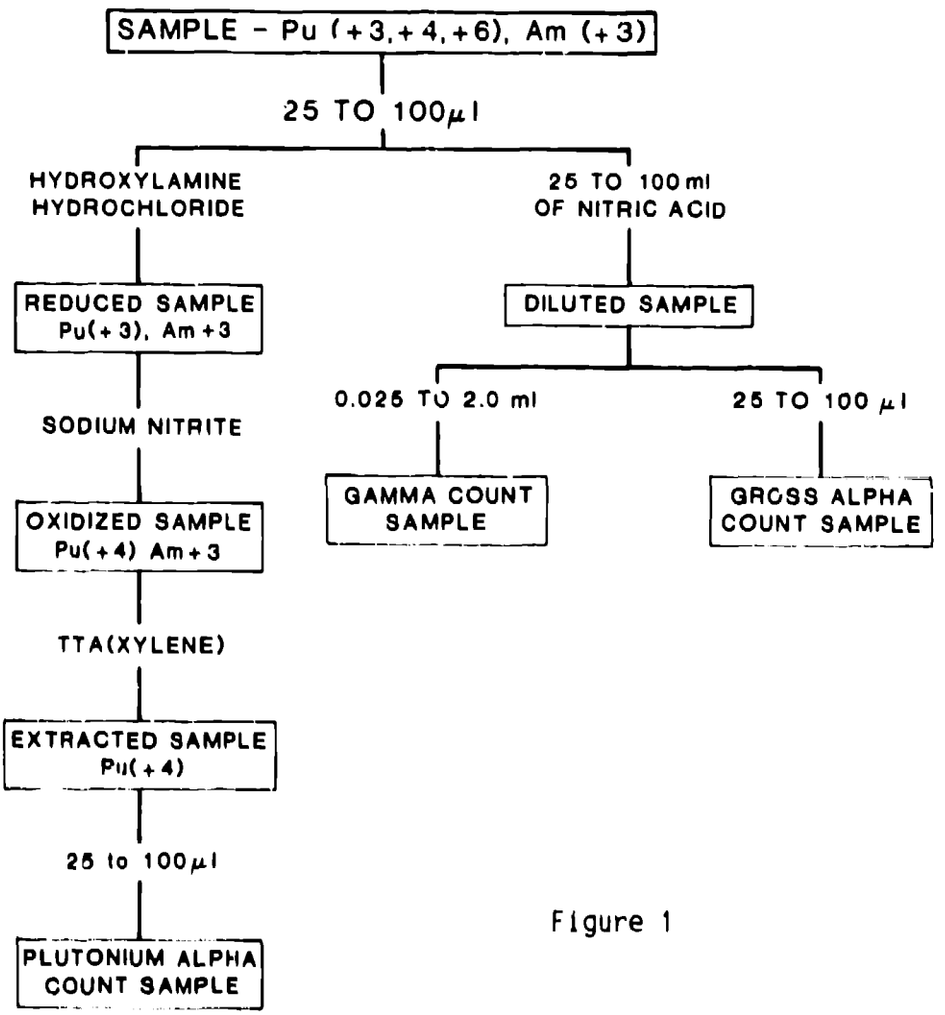

Figure 1

AUTOMATION OF THE PROCEDURES

An additional step was added in the dilution and extraction procedures when they were automated by the robotic system. An initial 1:10 dilution step allows the pipetting of larger aliquots throughout the procedures with the goal of increasing accuracy and precision.

A Zymate robotic system and associated unit modules were assembled and programmed to automate the procedures. The robot system consists of a controller, the robot arm, and six interchangeable hands. Two of the hands have syringes for pipetting, two hands have grippers for holding test tubes, etc., and two hands are dual function with each hand equipped with a set of grippers and a pipetting syringe. Unit modules include a centrifuge, a dry bath, two vortex mixers, a vial capping station, a shaker table, a balance, a hot plate, two peristaltic pumps, and two master laboratory stations each equipped with three syringes for dispensing reagents at two fixed and two moveable dispensers. The system has several racks for test tubes, pipet tips and cups.

The system is controlled by 150 programs that have been entered into the controller and that occupy almost all of its 64k memory. To facilitate the entering of sample information, an HP-85 computer was interfaced to the controller. The information is entered in an interactive mode on the HP-85 and is stored internally in a two-dimensional array. Once entered, the data are sent by columns to the robot where they are stored in a series of one-dimensional arrays.

RESULTS AND DISCUSSION

The fundamental operation that determines the accuracy and precision of the robotic sample preparation procedure is pipetting. If aliquots can be taken accurately and precisely, then the system can be made to perform as designed. In anticipation of this fact, a program was written for calibration of the various syringe hands by weighing 20 aliquots of water at 10, 30, 60, and 90% of the syringe volume. The mean values of the points are fitted to a linear equation by least squares. The calibration curve is stored in the controller and corrects pipetting operations. Results of running samples indicate a bias in the results for both plutonium and americium (Table 1). Therefore, the delivery of the syringes was checked by stepping through the actual sample preparation procedure with 20 samples. At each point where solution was pipetted, weights were taken and the volumes calculated. These results gave calibration curves that differed from those of the automatic calibration program. The following equations are an example:

(1) $y = 1.020x - 0.005$ (automatic)
(2) $y = 1.042x - 0.027$ (manual)

where: y = delivered volume
x = desired volume

At present the reason for the approximate 2% relative difference is not clear and further work will be done to resolve the problem.

In both calibration procedures, the reproducibility of the syringes is <1% RSD at a particular volume, and is generally closer to 1/2% RSD. An error analysis based on pipetting errors gave only an RSD $\leq$ 1% for the overall americium preparation and an RSD $\leq$ 2% for the overall plutonium preparation by dilution with this kind of pipetting precision. Establishing accurate calibration curves is the primary concern. Once this has been accomplished, analysis for plutonium and americium should be accurate to <5% at the 95% confidence level.

TABLE 1

COMPARISON OF MANUAL AND ROBOT SAMPLE PREPARATION FOR PLUTONIUM AND AMERICIUM ANALYSIS

Plutonium - *SD = 2.39% **Bias = -1.01%

Americium - *SD = 1.99% **Bias = 2.86%

*The SD is the standard deviation of the relative differences between the robotic value and the manual value for 24 analyses.

**The bias is the mean of the relative differences.

SUMMARY

A Zymate robotic system has been assembled and programmed to prepare samples for plutonium and americium analysis by radioactivity assay. The system has been checked and analysis of samples has started. Initial results of analyses are biased approximately 1% low for plutonium and approximately 3% high for americium compared with the manual method. The precision for both analyses is approximately 2.5% RSD. To determine the source of the bias and to improve precision, a systematic study of the pipetting operations has been started.

REFERENCES

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