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DETERMINATION OF NEODYMIUM-148 IN IRRADIATED URANIUM AND PLUTONIUM AS A MEASURE OF BURNUP

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GENERAL ELECTRIC
PLEASANTON, CALIFORNIA

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FOREWORD

The accurate determination of burnup in nuclear power plant fuels has called for the use of the most sensitive and precise tools available. The accurate isotopic dilution mass spectrometric technique has been applied to the determination of burnup by the nonradioactive refractory fission product Nd-148-to-fuel ratio method.

This report is one of a series of three reports that contain a summary of five years of work on accurate nuclear fuel burnup analysis. The other two reports are GEAP-5355 "BURNUP: A FORTRAN IV Code for Computing U and Pu Fuel Burnup U, Pu, and Nd Mass Spectrometric Measurements" and GEAP-5356 "A Survey and Evaluation of Thermal Fission Yields for U-235, Pu-239, U-233, and Pu-241."

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ABSTRACT

Nonradioactive fission product Nd^{148} is determined by isotopic dilution mass spectrometry. The isotopic diluent is a blend of Nd^{150}, U^{233}. and Pu^{242}. After an ion exchange separation of Pu, U, and Nd, each fraction is mounted on a Re filament for mass spectrometric analysis. Burnup (atom percent fission) is computed from the Nd^{148}-to-fuel ratio to an accuracy of 1.2% (1 σ) which includes a 0.7% single instrument uncertainty in the Nd^{148}-to-fuel ratio and a 1.0% uncertainty in the Nd^{148} fission yield.

INTRODUCTION

Several methods for the determination of burnup in nuclear fuels were investigated in this laboratory in 1960 to select the one best suited to determine whether a fuel had produced a warranted number of megawatt days per ton. It was reported[1] that both the Cs^{137}-to-uranium ratio method and the method based upon changes in the heavy element isotopic analysis had serious drawbacks. Since neither of these methods is adequate to determine power reactor fuel burnup at warranted levels in excess of 20,000 MWd/ton for recycled fuel to an accuracy of ± 1% deemed vital for multimillion dollar fuel warranty settlements, a program was undertaken to develop burnup methods having the required accuracy.

A review of all fission yields for U^{235} and Pu^{239} in thermal and fission neutron spectra reveals two mass regions of relatively constant fission yield.[2] These regions center at $Mo(Tc)^{99}$ and Nd^{148}. Colorimetric methods for the determination of Tc^{99} in reactor fuels have been published recently by other investigators.[3, 4] The thermal neutron absorption cross section for Tc^{99} is somewhat higher than for other commonly used burnup monitors. The usefulness of Tc^{99} as a burnup monitor may be limited to fuels operated below 1400°C. Above this temperature, Tc^{99} together with Mo, Ru, Rh, and Pd migrate to grain boundaries in oxide and carbide fuels and coalesce into acid-insoluble metallic inclusions under conditions which Nd and the rare earths appear to remain in acid-soluble solid solution. Mass spectrometric determination of molybdenum isotopes has been proposed, but there is some evidence of migration of MoO_3 in oxide fuels at temperatures to 2000°C common in these fuels.[5] Some patience and special techniques are required to obtain a good molybdenum ion beam, since there is a tendency for MoO_3 to sublime from the mass spectrometer filament before ionization temperatures are reached.[6]

The remaining region of interest near mass-148 is spanned by neodymium. First proposed as burnup monitor by Fudge, et al.,[7] Nd^{148} is not radioactive, requires no decay corrections and can be determined by the isotope dilution mass spectrometric method to the required accuracy of better than ± 1%.[8] Neodymium is not volatile, does not migrate in solid fuels below their recrystallization temperature,[9, 10] and has no volatile precursors. Neodymium-148 has a low destruction cross section, a low probability of formation from adjacent mass chains, and has good emission characteristics for mass analysis. Finally, Nd has a shielded isotope, Nd^{142} which can be used for correcting for natural Nd contamination and is not a normal constituent of unirradiated fuel.

NEODYMIUM-148 METHOD DEVELOPMENT

Recent work in this laboratory has been concentrated on the development of Nd^{148} as an indicator of total fissions occurring in a nuclear fuel. The low specific activity of the Nd fraction[11] and the small sample sizes required for mass analysis[12] enable burnup to be measured without the use of an elaborate shielded facility. It was established that Nd^{148} has a low thermal neutron cross section of 2.54 ± 0.18 barns[13] and that its resonance integral is only 18.7 ± 1.5 barns.[13] As a result, burnout corrections are negligibly small even at the highest exposures expected for power reactor fuels. The world average Nd^{148} yields in thermal fission of U^{235}, U^{233}, and Pu^{239} are 1.66, 1.29, and 1.65%, respectively.[14-17] The epithermal yield of Nd^{148} in the same three fissionable nuclides does not appreciably differ from that in thermal fission. The yield[17] of Nd^{148} for thermal fission of Pu^{241} is 1.84% and for fast fission of U^{238} is 1.97%.

A chemical separation and mass spectrometric procedure for the determination of Nd^{148} in irradiated uranium fuel was written for the American Society for Testing and Materials with ASTM Designation E-321. This method[18] makes use of a blend of three enriched isotopes, Nd^{150}, U^{233}, and Pu^{242} as isotopic diluents. An inter-laboratory study of this method[16] showed a multi-laboratory coefficient of variation (one standard deviation) of 0.9% for Nd^{148} concentration and ± 1.2% for uranium concentration. Where greatest possible precision and accuracy are required, such as for fuel warranty settlements, it is recommended that measurements be based on ASTM method E-321 because of its small multi-laboratory coefficient of variation. The step-by-step procedure appears in Appendix A. In the preparation of this procedure, an attempt was made to simplify the chemistry and the calculations to attain the wide acceptance and use that this method deserves.

An earlier procedure[19] had somewhat greater versatility and achieved somewhat greater decontamination, however, it was not sufficiently short and simple to be generally acceptable. Significant among the changes made in neodymium chemistry are the elimination of (a) the unconventional reversed phase chromatography, which required preparation of a special matrix, and (b) the final cation exchange chromatography which required the cumbersome preparation of 12 M HCl in aqueous ethanol with gaseous HCl. A preliminary anion exchange separation of a fission product fraction (including neodymium) from the major fuel atoms is included because in large concentrations of other atoms, the neodymium band is broadened and the neodymium elution position altered during the purification of neodymium by anion exchange chromatography.

To obtain good resolution in anion exchange chromatography, and therefore clean separations, adequate time must be allowed to maintain equilibrium conditions. For this reason, flow rates are kept below 1 cm/min. Because of the low boiling point of methanol, high-temperature operation cannot be used to improve the reaction kinetics.

The sample size recommended in the neodymium procedure is large enough to minimize the effect of a natural neodymium blank. In the majority of samples the blank averages 0.7 nanogram of Nd^{148} largely from the rhenium filament. A 70-nanogram sample size which is 100 times greater than this blank is considered adequate because a correction for this blank is included in the calculations. Deletion of the HCl from the filament mounting solution has acted further to reduce the blank by minimizing filament etching.

Since the same amount of fission products is taken for each analysis, the radiation dose from each sample is relatively constant for all burnup levels and depends principally upon the length of time since irradiation. Gamma dose rates vary from 10 mR/h at 1 meter for samples of 60-day cooled fuel to 1 mR/h at 1 meter for samples of 1-year cooled fuels. Beta dose rates are an order of magnitude greater but can be effectively

stopped with a 1/2-inch-thick transparent plastic sheet. By the use of such simple local shielding, dilute solutions of irradiated nuclear fuel dissolver solution can be analyzed for burnup without an elaborately shielded analytical facility.

Other significant changes in the neodymium chemical separation include a chloride removal step before the anion exchange separation of the neodymium nitrate complex. Any chlorides in the dissolver solution (e. g., from aqua regia dissolution) otherwise prevent the absorption of the neodymium nitrate complex. A tracer Nd^{147} run is recommended for each new lot of resin to establish its suitability in this analysis.

To establish the multi-laboratory precision of the method, round robin samples were distributed to eight other laboratories for analysis. Adoption of a single-blended Nd^{150}, Pu^{242}, and U^{233} spike should improve the accuracy of the atom per cent fission measurement since the actual volumes of both sample and spike cancel and the method relies only on the measurement of ratios. The inclusion of a mass discrimination correction should remove any ratio biases among laboratories.

This laboratory has shown the Nd^{148} can be analyzed with great precision (± 0. 8%). However, because this simplified procedure for Nd^{148} (Appendix A) requires less time and materials than a radiochemical Cs^{137} analysis, a Nd^{148} analysis can also be quicker and cheaper than a Cs^{137} analysis for a laboratory that has a mass spectrometer. Fortunately, most nuclear fuel reprocessing plants and atomic energy laboratories already have a mass spectrometer for the determination of isotopic abundances in uranium.

Burnup (atom percent fission) is computed from the Nd^{148}-to-fuel ratio to an accuracy of 1. 2% (1 σ) which includes a 0. 7% single instrument uncertainty in the Nd^{148}-to-fuel ratio (see Table I, page A-14) and a 1. 0% uncertainty in the Nd^{148} fission yield.

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APPENDIX A

DETERMINATION OF ATOM PER CENT FISSION IN URANIUM AND PLUTONIUM FUEL

Stable Fission Product Nd^{148} Method

1. SCOPE

1.1 This method covers the determination of stable fission product Nd^{148} in irradiated U fuel (with initial Pu content from 0 to 50 per cent) as a measure of fuel burnup (1-3). [1]

2. SUMMARY OF METHOD

2.1 Fission product Nd is chemically separated from irradiated fuel and determined by isotopic dilution mass spectrometry. Enriched Nd^{150} is selected as the Nd isotope diluent, and the mass ^{142} position is used to monitor for natural Nd contamination. The two rare earths immediately adjacent to Nd do not interfere. Interference from other rare earths, such as natural or fission product Ce^{142} or natural Sm^{148} and Sm^{150} is avoided by removing them in the chemical purification (4).

2.2 After addition of a blended Nd^{150}, U^{233}, and Pu^{242} spike to the sample, the Nd, U, and Pu fractions are separated from each other by ion exchange. Each fraction is further purified for mass analysis.

2.3 The gross alpha, beta, and gamma decontamination factors are in excess of 10^3 and are normally limited to that value by traces of Cm^{242}, Pm^{147}, and Am^{241}, respectively, (and sometimes Ru^{106}) none of which interfere in the analysis. The 70 nanogram Nd^{148} minimum sample size recommended in the procedure is large enough to exceed by 100-fold a typical natural Nd blank of 0.7 ± 0.7 nanogram Nd^{148} (for which a correction is made) without exceeding radiation dose rates of 20 mR/h at 1 meter. Since a constant amount of fission products are taken for each analysis, the radiation dose from each sample is similar for all burnup values and depends principally upon cooling time. Gamma dose rates vary from 20 mR/h at 1 meter for 60-day cooled fuel to 2 mR/h at 1 meter for 1-year cooled fuel. Beta dose rates are an order of magnitude greater, but can be shielded out with a 1/2-inch-thick plastic sheet. By use of such simple local shielding, dilute solutions of irradiated nuclear fuel dissolver solutions can be analyzed for burnup without an elaborate shielded analytical facility. The decontaminated Nd fraction is mounted on a Re filament for mass analysis. Samples from 20 nanogram to 20 μg run well in the mass spectrometer with both NdO^+ and Nd^+ ion beams present. The metal ion is enhanced by deposition of carbonaceous material on the filament as oxygen getter.

SIGNIFICANCE

3.1 The burnup of an irradiated nuclear fuel can be determined from the amount of a fission product formed during irradiation. Among the fission products, Nd^{148} has the following properties to recommend it as an ideal burnup indicator: (a) It is not volatile, does not migrate in solid fuels below their recrystallization temperature, and has no volatile precursors. (b) It is nonradioactive and requires no decay corrections. (c) It has a low destruction cross section and a low formation cross section from adjacent mass chains. (d) It has good emission characteristics for mass analysis. (e) Its fission yield is nearly the same for U^{235} and Pu^{239} and is essentially independent of neutron energy (5). (f) It has a shielded isotope, Nd^{142}, which can be used for correcting natural Nd contamination. (g) It is not a normal constituent of unirradiated fuel.

3.2 The analysis of Nd^{148} in irradiated fuel does not depend on the availability of pre-irradiation sample data or irradiation history. Atom percent fission is directly proportional to the Nd^{148}-to-fuel ratio in irradiated fuel.

3.3 The method can be applied directly to uranium fuel containing less than 0.5 per cent initial Pu with 1 to 100 gigawatt days per tonne burnup. For fuel containing 5 to 50 per cent initial Pu, increase the Pu content by a factor of 10 to 100, respectively in both reagents 4.3 and 4.4.

4. REAGENTS AND MATERIALS

4.1 Purity of Reagents - Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society.[2] Other reagents may be used, provided they are of sufficient purity to give the same accuracy.

4.2 Purity of Water - Unless otherwise indicated, water used shall conform to the Specifications for Reagent Water (ASTM Designation: D 1193).[3]

4.3 Blended Nd^{148}, Pu^{239}, and U^{238} Calibration Standard - Prepare a solution containing about 0.0400 mg Nd^{148}/liter, 50 mg U^{238}/liter, 2.5 mg Pu^{239}/liter, in (1:1) HNO_3 with 0.01 M HF as follows. With a new calibrated, clean, Kirk-type micropipet, add 0.500 ml of Pu^{239} known solution (see 4.18) to a calibrated 1-liter volumetric flask. Rinse the micropipet into the flask three times with (1:1) HNO_3. In a similar manner, add 0.500 ml of U^{238} known solution (see 4.20) and 1.000 ml of Nd^{148} known solution (see 4.15). Add 10 drops of concentrated HF and dilute exactly to the 1-liter mark with (1:1) HNO_3 and mix thoroughly.

4.3.1 From K_{148} (see 4.15), calculate the atoms of Nd^{148}/ml of calibration standard, C_{148}, from

$$C_{148} = \frac{\text{ml } Nd^{148} \text{ known solution}}{1000 \text{ ml calibration standard}} \times K_{148}$$

4.3.2 From K_{238} (see 4.20), calculate the atoms of U^{238}/ml of calibration standard, C_{238}, from

$$C_{238} = \frac{\text{ml } U^{238} \text{ known solution}}{1000 \text{ ml calibration standard}} \times K_{238}$$

4.3.3 From K_{239} (see 4.18), calculate the atoms of Pu^{239}/ml of calibration standard, C_{239}, from

$$C_{239} = \frac{\text{ml } Pu^{239} \text{ known solution}}{1000 \text{ ml calibration standard}} \times K_{239}$$

4.3.4 Flame-seal 3- to 5-ml portions in glass ampoules to prevent evaporation after preparation until time of use. For use, break off the tip of an ampoule, pipet promptly the amount required, and discard any unused solution. If more convenient, calibration solution can be flame-sealed in pre-measured 1000 μliter portions for quantitative transfer when needed.

4.4 Blended Nd^{150}, U^{233}, and Pu^{242} Spike Solution - Prepare a solution containing about 0.4 mg Nd^{150}/liter, 50 mg U^{233}/liter, and 2.5 mg Pu^{242}/liter in (1:1) HNO_3 with 0.01 M HF. These isotopes are obtained in greater than 95, 99, and 99 percent isotopic purity, respectively, from the Isotopes Sales Department of Oak Ridge National Laboratory. Standardize the spike solution as follows:

4.4.1 In a 5-ml beaker, place about 0.1 ml of ferrous solution, exactly 500 μliter of calibration standard (see 4.3) and exactly 500 μliter of spike solution (see 4.4). In a second beaker, place about 0.1 ml of ferrous solution and 1 ml of calibration standard without any spike. In a third beaker, place about 0.1 ml of ferrous solution and 1 ml of spike solution without standard. Mix well and allow to stand for 5 min to reduce Pu (VI) to Pu (III) or Pu (IV).

4.4.2 Follow the procedure described in 6.2.3 through 6.5.5. On the Pu fractions, record the atom ratios of Pu^{242} to Pu^{239} in the calibration standard, $C_{2/9}$; in the spike solution, $S_{2/9}$; and in the standard-plus-spike mixture, $M_{2/9}$. On the U fractions record the corresponding U^{233}-to-U^{238} ratios, $C_{3/8}$, $S_{3/8}$, and $M_{3/8}$. On the Nd fractions, record the corresponding Nd^{150}-to-Nd^{148} ratios, $C_{50/48}$, $S_{50/48}$, and $M_{50/48}$. Correct all average measured ratios for mass discrimination bias (see 5.2).

4.4.3 Calculate the number of atoms of Nd^{150}/ml of spike, A_{50}, from

$$A_{50} = C_{148} \left(\frac{M_{50/48} - C_{50/48}}{1 - M_{50/48}/S_{50/48}} \right)$$

4.4.4 Calculate the number of atoms of U^{233}/ml of spike, A_{33}, from

$$A_{33} = C_{238} \left(\frac{M_{3/8} - C_{3/8}}{1 - M_{3/8}/S_{3/8}} \right)$$

4.4.5 Calculate the number of atoms of Pu^{242}/ml spike, A_{42}, from

$$A_{42} = C_{239}\left(\frac{M_{2/9} - C_{2/9}}{1 - M_{2/9}/S_{2/9}}\right)$$

4.4.6 Store in the same manner as the calibration standard (see 4.3). that is, flame-seal 3-to 5-ml portions in glass ampoules. For use, break off the tip of an ampoule, pipet promptly the amount required and discard any unused solution. If more convenient, spike solution can be flame-sealed in a pre-measured 1000 μliter portions for quantitative transfer to individual samples.

4.5 Dowex 1 Resin - Convert Dowex 1- × 2 or 1-× 4 (200-400 mesh) chloride-form resin[4] to nitrate form by washing 200 ml of resin in a suitable column (for example, a 250-ml buret) with (1:1) HNO_3 until a drop of effluent falling into an $AgNO_3$ solution remains clear. Finally, rinse with water, and dry overnight in a vacuum dessicator. Store the resin in an airtight container. Since the elution characteristics of ion exchange resins depend upon their actual percentage cross linkage and particle size (surface-to-volume ratio), which may vary from one lot to the next, it is most convenient to set aside a bottle of resin to be used solely for this procedure. Before use on actual samples, a small amount of tracer Nd^{147} should be taken through the procedure. Collect each consecutive 8-cm fraction of eluant and count for γ radioactivity. If over 80 per cent of the Nd^{147} appears in the Nd fraction, the resin can be used as directed; if not, small adjustments can be made in the elution volumes collected.

4.6 Ferrous Solution (0.001 M) - Add 40 mg of reagent grade ferrous ammonium sulfate ($Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$) and 1 drop of concentrated H_2SO_4 to 5 ml of redistilled water. Dilute to 100 ml with water and mix. This solution does not keep well. Prepare fresh daily.

4.7 Filament Mounting Solution - Dissolve 70 mg sucrose in 100 ml of water.

4.8 Hydrochloric Acid - Prepare reagent low in uranium and dissolved solids by saturating redistilled water in a polyethylene container to 12 M with HCl gas which has passed through a quartz-wool filter. Dilute 1:1 and 1:24 with redistilled water. Store each solution in a polyethylene container. One drop of acid, when evaporated on a clean microscope slide cover glass, must leave no visible residue. Test monthly. Commercial HCl (CP grade) in glass containers has been found to contain excessive residue (dissolved glass) which inhibits ion emission.

4.9 Hydrofluoric Acid - Reagent grade concentrated HF (28M).

4.10 Ion Exchange Column - One method of preparing such a column is to draw out the end of a 15-cm (6 inches) length of 4-mm inside diameter glass tubing and force a glass wool plug into the tip tightly enough to restrict the linear flow rate of the finished column to less than 1 cm/min. By means of a capillary pipet or polyethylene wash bottle add resin (see 4.5) suspended in loading solution (see 4.13) to the required bed length. Since the diameter of glass tubing may vary from one laboratory to another, the quantity of resin and the quantity of liquid reagents used are specified in centimeters of column length.

4. 10. 1 To simplify use, mark the tubing above the resin bed in centimeters with a marking pen or back it with a strip of centimeter graph paper. Dispense liquid reagents into the column from a polyethylene wash bottle to the length specified in the procedure. Thus, 160 cm of wash solution can be added by filling to the 8 cm mark twenty times. However, it is more convenient to measure this 160 cm of wash solution by placing a 6 dram vial under the column. Mark the vial at the liquid level reached by 160 cm of wash solution. Wash other columns of the same diameter until the same liquid level is reached in the vial.

4. 10. 2 The efficiency of an ion exchange separation is always limited by the kinetics of the system. A non-aqueous system such as methanolic nitric acid has slower kinetics than an aqueous system. Elevated temperature operation (which would improve kinetics) cannot be resorted to because of the low boiling point of methanol. An adequate separation of neodymium from americium at room temperature requires a column residence time of 180 min. Column operating parameters have been chosen to obtain this residence time.

4. 11 Methanol - Absolute.

4. 12 Methanolic HNO_3 Eluant - Pipet 10 ml of (1:500) nitric acid into a 100-ml volumetric flask and dilute to the mark with absolute methanol. Protect this reagent against preferential evaporation of methanol by keeping it in a polyethylene wash bottle. Prepare fresh weekly.

4. 13 Methanolic HNO_3 Loading Solution - Pipet 1 ml of (1:1) HNO_3 into a 10-ml volumetric flask and dilute to the mark with absolute methanol. Store as 4. 12. Prepare fresh weekly. High nitrate loading solution is used to ensure absorption of Nd in a tight band and to overcome interference from sulfate and fluoride ions.

4. 14 Methanolic HNO_3 Wash Solution - Pipet 10 ml of (1:100) HNO_3 into a 100-ml volumetric flask and dilute to the mark with absolute methanol. Store as 4. 12. Prepare fresh weekly.

4. 15 Nd^{148} Known Solution - Heat natural Nd_2O_3 (> 99. 9 per cent pure) in an open crucible at 900 C for 1 hour to destroy any carbonates present and cool in a dessicator. Weigh 0. 4071 gm Nd_2O_3 and place it in a calibrated 500-ml volumetric flask. Dissolve the oxide in (1:1) HNO_3 and dilute to the 500-ml mark with (1:1) HNO_3 and mix thoroughly. By using the weight of Nd_2O_3 in grams, and the purity, calculate the atoms of Nd^{148}/ml of known solution, K_{148}, from

$$K_{148} = \frac{\text{g } Nd_2O_3}{500 \text{ ml}} \times \frac{\% \text{ purity}}{100} \times \frac{50.38 \text{ mg } Nd^{148}}{1 \text{ g } Nd_2O_3} \times \frac{6.025 \times 10^{20} \text{atoms}}{147.92 \text{ molecular weight}}$$

4.16 Nitric Acid - (1:1, 1:5, 1:30, 1:100 and 1:500) Prepare by diluting 1 part of concentrated (16 M) nitric acid with 1, 5, 30, 100, and 500 parts of water, respectively.

4.17 Perchloric acid - 70 per cent $HClO_4$.

4.18 Pu^{239} Known Solution. - Add 10 ml of (1:1) HCl to a clean calibrated 100-ml flask. Cool the flask in an ice water bath. Allow time for the acid to reach approximately 0 C and place the flask in a glove box. Displace the air in the flask with inert gas (Ar, He, or N_2). Within the glove box, open the U. S. National Bureau of Standards Plutonium Metal Standard Sample 949, containing about 0.5 g Pu (actual weight individually certified), and add the metal to the cooled HCl. After dissolution of the metal is complete, add 1 drop of concentrated HF and 40 ml of (1:1) HNO_3 and swirl. Place the flask in a stainless steel breaker for protection and invert a 50-ml beaker over the top and let it stand for at least 8 days to allow any gaseous oxidation products to escape. Dilute to the mark with (1:1) HNO_3 and mix thoroughly. By using the individual weight of Pu in grams, the purity, and the molecular weight of the Pu given on the NBS certificate, with the atom fraction, A_9, determined as in 7.8, calculate the atoms of Pu^{239}/ml of Pu^{239} known solution, K_{239}, from

$$K_{239} = \frac{\text{mg Pu}}{\text{100 ml solution}} \times \frac{\text{\% purity}}{100} \times \frac{6.025 \times 10^{20}\ \text{atoms}}{\text{Pu molecular weight}} \times A_9$$

4.19 Sodium Nitrite Solution (0.1 M) - Add 0.69 g reagent grade sodium nitrite ($NaNO_2$) and 0.2 g NaOH to 50 ml redistilled water, dilute to 100 ml with redistilled water and mix.

4.20 U^{238} Known Solution - Heat U_3O_8 from the National Bureau of Standards Natural Uranium Oxide Standard Sample 950 in an open crucible at 900 C for 1 hour and cool in a dessicator in accordance with the certificate accompanying the standard sample. Weigh about 12.0 g U_3O_8 accurately to 0.1 mg and place it in a calibrated 100-ml volumetric flask. Dissolve the oxide in (1:1) HNO_3. Dilute to the 100-ml mark with (1:1) HNO_3 and mix thoroughly. By using the measured weight of U_3O_8 in grams, the purity given on the NBS certificate, and the atom fraction U^{238}, A_8, determined as in 7.5, calculate the atoms U^{238}/ml of U^{238} solution, K_{238}, from

$$K_{238} = \frac{\text{g } U_3O_8}{\text{100 ml solution}} \times \frac{\text{\% purity}}{100} \times \frac{848.0\ \text{mg U}}{1\ \text{g } U_3O_8} \times \frac{6.025 \times 10^{20}\ \text{atoms}}{238.03\ \text{molecular weight}} \times A_8$$

5. INSTRUMENT CALIBRATION

5.1 In the calibration of the mass spectrometer for the analysis of neodymium, uranium, and plutonium, the measurement and correction of mass discrimination bias is an important factor in obtaining accurate and consistent results. The mass discrimination bias can be readily measured on natural neodymium where the Nd^{142}-to-Nd^{150} ratio spans over a 5 per cent spread in mass. The mass discrimination bias factor, B, is constant for Nd, U, and Pu analysis for a given method of scanning (e. g., by varying either acceleration voltage or magnetic field) and for a given method of detection (e. g., by pulse counting or current integration) on a given detector (e. g., electron multiplier, scintillation detector, or D. C. collector plate). Calculate B from

$$B = \frac{1}{c}\left(\frac{\bar{R}_{i/j}}{R_s} - 1\right)$$

where:

$\bar{R}_{i/j}$ = average measured atom ratio of isotope i to isotope j. For the most accurate determination of B, let $\bar{R}_{i/j}$ be the average measured atom ratio of Nd^{142} to Nd^{150},

R_s = known value of the measured atom ratio. For the ratio of Nd^{142} to Nd^{150} in natural neodymium, $R_s = 4.824$, and

c = Δ mass/mass. The value of c for various ratios and ion species include

Ratio	Nd^+, U^+, or Pu^+	NdO^+, UO_2^+, or PuO_2^+
Nd^{148}/Nd^{150}	$+\frac{2}{150}$	$+\frac{2}{166}$
Nd^{150}/Nd^{148}	$-\frac{2}{148}$	$-\frac{2}{164}$
Nd^{142}/Nd^{150}	$+\frac{8}{150}$	$+\frac{8}{166}$
U^{234}/U^{238}	$+\frac{4}{238}$	$+\frac{4}{270}$
U^{235}/U^{238}	$+\frac{3}{238}$	$+\frac{3}{270}$
U^{236}/U^{238}	$+\frac{2}{238}$	$+\frac{2}{270}$
U^{238}/U^{233}	$-\frac{5}{233}$	$-\frac{5}{265}$
U^{233}/U^{238}	$+\frac{5}{238}$	$+\frac{5}{270}$
Pu^{240}/Pu^{239}	$-\frac{1}{239}$	$-\frac{1}{271}$
Pu^{241}/Pu^{239}	$-\frac{2}{239}$	$-\frac{2}{271}$
Pu^{242}/Pu^{239}	$-\frac{3}{239}$	$-\frac{3}{271}$

5.2 Correct every measured average ratio, $\bar{R}_{i/j}$, for mass discrimination as follows:

$$R_{i/j} = \frac{\bar{R}_{i/j}}{(1 + cB)}$$

where:

$R_{i/j}$ = the corrected average atom ratio of isotope i to isotope j.

6. PROCEDURE

6.1 Preparation of a Working Dilution of Dissolver Solution:

6.1.1 Prepare a dilution of fuel dissolver solution with (1:1) nitric acid to obtain a concentration of 100 to 1000 mg U plus Pu/liter.

6.2 Preliminary Neodymium, Uranium, and Plutonium Separation:

6.2.1 In a 10-ml beaker, place 1000 μliters of spike solution (see 4.4) and an aliquot of sample containing about 70 nanogram of fission product Nd^{148}. In a second beaker, place a similar aliquot of sample without any spike solution. If the approximate burnup in gigawatt days per tonne is known, the number of milligrams of U plus Pu required for the analysis can be read from Figure A-1. Follow the remaining procedure on each solution.

6.2.2 Add about 0.1 ml of ferrous solution (see 4.6). Mix well and allow to stand for 5 min to reduce Pu (VI) to Pu (III) or Pu (IV).

6.2.3 Add one drop (20 to 50 μliters) of nitrite solution (see 4.19) to oxidize all plutonium to the tetravalent state and evaporate to near dryness to reduce volume. Dissolve the residue in 250 μliter of (1:1) nitric acid; take care to ensure complete dissolution.

6.2.4 Prepare a 2-cm long anion exchange column (see 4.10) for operation at 50 to 60 C. Wash the column with 10 cm of (1:500) nitric acid followed by 10 cm of (1:1) nitric acid. Place a clean 5-ml beaker under the column.

6.2.5 Transfer the sample solution onto the column with a disposable capillary pipet. Carefully wash down walls of the column with a few drops of (1:1) nitric acid to ensure all the sample is adsorbed on the column.

6.2.6 Elute the neodymium into the 5-ml beaker with 5 cm of (1:1) nitric acid. Purify this neodymium solution by the procedure given in 6.3.

6.2.7 Elute the uranium into a second 5-ml beaker with 20 cm of (1:5) nitric acid. Purify this uranium solution by the procedure given in 6.4

6.2.8 Wash the column with 50 cm of (1:5) nitric acid. Discard this wash. Elute the plutonium with 20 cm of (1:30) nitric acid into a third 5-ml beaker. Purify this plutonium solution by the procedure given in 6.5.

6.3 Neodymium Purification:

6.3.1 Evaporate the neodymium solution from 6.2.6 to near dryness. Dissolve the residue in 500 μliter of loading solution (see 4.13).

6.3.2 Prepare a 2-cm long anion exchange column (see 4.10) for room temperature operation. Transfer the neodymium solution onto the column with a disposable capillary pipet. Wash down the walls of the column with a few drops of loading solution (see 4.13) to ensure all the neodymium is adsorbed on the column.

6.3.3 Elute the column with eluant (see 4.12). Discard the first 8 cm of eluant. Collect in a 5-ml beaker the next 32-cm of eluant containing the neodymium. Evaporate the solution to dryness.

6.3.4 Redissolve the residue in 500 μliters of (1:1) nitric acid. Add 1 ml of 70 per cent $HClO_4$ and again evaporate to dryness. Redissolve the residue in about 500 μliters of loading solution (see 4.13).

6.3.5 Prepare a 6-cm-long ion exchange column (see 4.10). Transfer the sample solution into the column with a capillary pipet. Rinse the sample beaker into the column with several drops of loading solution. Finally, rinse down the walls of the column with a few drops of loading solution to ensure that all the sample is adsorbed in the resin.

6.3.6 Pass 160 cm of wash solution (see 4.14) through the column. This amount of wash solution is just sufficient to elute the rare earth elements (Pm and heavier) and the actinide elements (Am and heavier) as shown in Figure A-2.

6.3.7 Strip the Nd from the column with 32 cm (see Fig. A-2) of eluant (see 4.12). Collect the Nd solution in a 5-ml centrifuge tube. Evaporate this solution to dryness in a hot water bath with a gentle stream of filtered air.

6.3.8 Dissolve the Nd in a small drop of filament mounting solution and evaporate it onto a Re filament for mass spectrometry.

6.3.9 Measure the Nd^{148}-to-Nd^{150} and the Nd^{142}-to-Nd^{150} atom ratio for each prepared filament by means of a surface ionization mass spectrometer. Correct each average measured ratio for mass discrimination bias (see 5.2).

6.4 Uranium Purification:

6.4.1 Evaporate the uranium solution (see 6.2.7) to dryness. Add a few drops of 12 M HCl and again evaporate to dryness.

6.4.2 Prepare a 5-mm long anion exchange column (see 4.10) for operation at 50 to 60 C. Wash the column with 10 cm of (1:24) HCl and 10 cm of (1:1) HCl.

6.4.3 Redissolve the uranium in 500 μliters of (1:1) HCl and transfer it to the column. Wash the column with 15 cm of (1:1) HCl. Discard this wash.

6.4.4 Elute the uranium with 5 cm of (1:24) HCl into a 5-ml centrifuge tube, and evaporate to dryness in a boiling water bath with a gentle stream of filtered air. Dissolve the uranium in 1 drop of Filament Mounting Solution (see 4.7) and evaporate it onto a rhenium filament for mass spectrometry.

6.4.5 Measure the U^{234}, U^{235}, and U^{236} to U^{238} atom ratio ($R_{4/8}$, $R_{5/8}$, and $R_{6/8}$) and the U^{238} to U^{233} atom ratio, $R_{8/3}$, on each unspiked uranium sample and the U^{238} to U^{233} atom ratio, $M_{8/3}$, on each spiked uranium sample by means of a surface ionization mass spectrometer. Correct each average measured ratio for mass discrimination (see 5.2).

6.5 Plutonium Purification:

6.5.1 To the plutonium solution (see 6.2.8) add 1 ml of concentrated nitric acid and evaporate to 100 μliter volume. Do not evaporate to dryness, which might thermally decompose the nitrate to oxide; such oxides are difficult to redissolve.

6.5.2 Prepare a 5-mm long anion exchange column (see 4.10) for operation at 50 to 60 C. Wash the column with 10 cm of (1:24) HCl followed by 10 cm of (1:1) HNO_3.

6.5.3 Dilute the plutonium with 5 drops of (1:1) HNO_3 and transfer it to the column with a disposable capillary pipet. Rinse the beaker with 5 drops of (1:1) HNO_3 and transfer the rinse to the column. Wash the column with 25 cm of (1:5) HNO_3. Discard this wash. Elute the plutonium with 5 cm of (1:24) HCl into a 5-ml centrifuge tube.

6.5.4 Evaporate the solution to dryness in a boiling water bath with a gentle stream of filtered air. Dissolve the plutonium in 1 drop of (1:24) HCl and evaporate it onto a rhenium filament by passing a small electrical current through the filament. Evaporate 1 drop of filament mounting solution (see 4.7) over the sample and increase the current briefly to char the sucrose from the mounting solution.

6.5.5 Measure the Pu^{240}, Pu^{241}, and Pu^{242} to Pu^{239} atom ratio ($R_{0/9}$, $R_{1/9}$ and $R_{2/9}$) on each unspiked plutonium sample and the Pu^{239} to Pu^{242} atom ratio, $M_{9/2}$, on each spiked plutonium sample by means of a surface ionization mass spectrometer. Correct each average measured ratio for mass discrimination (see 5.2).

7. CALCULATIONS

7.1 Calculate the ratio of effective fission yields of Nd^{150} to Nd^{148}, $E_{50/48}$ from

$$E_{50/48} = \frac{R_{50/48}\,(R_{50/42} - C_{50/42})}{R_{50/42} - R_{50/48}\,(C_{48/42})}$$

where:

$R_{50/48}$, $R_{50/42}$ = atom ratio of Nd^{150} to Nd^{148} and Nd^{150} to Nd^{142} in the unspiked sample, corrected for mass discrimination bias.

$C_{50/42}$, $C_{48/42}$ = atom ratios of Nd^{150} to Nd^{142} and Nd^{148} to Nd^{142} in natural neodymium contamination.

7.2 Calculate constants, a, b, c, d, e, and f from

$$a = C_{42/50} - S_{42/50} \quad (1)$$

$$b = C_{48/50} - S_{48/50} \quad (2)$$

$$c = C_{42/50}\,S_{48/50} - S_{42/50}\,C_{48/50} \quad (3)$$

$$d = C_{42/50} \quad (4)$$

$$e = E_{50/48}\,C_{42/50} \quad (5)$$

$$f = (1 - E_{50/48}\,C_{48/50}) \quad (6)$$

where:

$C_{42/50}$, $C_{48/50}$ = atom ratio of Nd^{142} and Nd^{148} to Nd^{150} in natural neodymium contamination, which are 4.824 and 1.0195, repsectively,

$S_{42/50}$, $S_{48/50}$ = atom ratio of Nd^{142} and Nd^{148} to Nd^{150} respectively in the spike solution.

7.3 Calculate $M'_{48/50}$ from

$$M'_{48/50} = \frac{a\,M_{48/50} - b\,M_{42/50} - c}{d - e\,M_{48/50} - f\,M_{42/50}}$$

where:

$M'_{48/50}$ = atom ratio of fission product Nd^{148} to spike Nd^{150} adjusted for fission product Nd^{150}, Nd^{148} impurity in Nd^{150} spike, and Nd^{148} and Nd^{150} from natural neodymium contamination.

$M_{48/50}$, $M_{42/50}$ = measured atom ratio of Nd^{148} to Nd^{150} and Nd^{142} to Nd^{150} of the sample plus spike mixture corrected for mass discrimination bias (see 5.2).

7.4 Calculate the number of fissions per sample, F', from

$$F' = \left(\frac{A_{50}}{E_{48}}\right) M'_{48/50}$$

where:

E_{48} = effective fractional fission yield of Nd^{148} calculated from the fission yields of Nd^{148} for each of the fissioning isotopes weighted according to their contribution to fission as measured in ASTM Method E-244. For U^{235} fuels, E_{48} can be assumed to be the fractional yield for Nd^{148} in U^{235} thermal fission which is 0.0166 [see Ref. (6)]; and

A_{50} = the number of atoms of Nd^{150}/ml of spike (see 4.4.3).

7.5 Calculate the atom fraction U^{238} in the unspiked uranium sample, A_8, from

$$A_8 = \frac{R_{8/8}}{R_{4/8} + R_{5/8} + R_{6/8} + R_{8/8}}$$

where $R_{8/8}$ (which equals 1) is retained for clarity.

7.6 Calculate $S_{8/3}$ from $S_{3/8}$ (see 4.4.2)

$$S_{8/3} = 1/S_{3/8}$$

7.7 Calculate the total uranium atoms per sample, U', from A_{33} (see 4.4.4)

$$U' = \frac{A_{33}}{A_8}\left(\frac{M_{8/3} - S_{8/3}}{1 - M_{8/3}/R_{8/3}}\right)$$

7.8 Calculate the atom fraction Pu^{239} in the unspiked plutonium sample, A_9, from

$$A_9 = \frac{R_{9/9}}{R_{9/9} + R_{0/9} + R_{1/9} + R_{2/9}}$$

where $R_{9/9}$ (which equals 1) is retained for clarity.

7.9 Calculate $S_{9/2}$ from $S_{2/9}$ (see 4.4.2) and $R_{9/2}$ from $R_{2/9}$

$$S_{9/2} = 1/S_{2/9}$$

$$R_{9/2} = 1/R_{2/9}$$

7.10 Calculate the total plutonium atoms per sample, Pu', from A_{42} (see 4.4.5)

$$Pu' = \frac{A_{42}}{A_9}\left(\frac{M_{9/2} - S_{9/2}}{1 - M_{9/2}/R_{9/2}}\right)$$

7.11 Calculate the total heavy element atom percent fission, F_T, from

$$F_T = \frac{F'}{U' + Pu' + F'} \times 100$$

7.12 If desired, calculate the gigawatt days per tonne from

$$\text{gigawatt days per tonne} = F_T \times (9.6 \pm 0.3).$$

8. PRECISION

8.1 The single-instrument precisions for the average of duplicate determinations of F′, U′, and F_T are given in Table 1 in percent relative (1S) as defined in ASTM Recommended Practice E 177. The expected average difference (average range) between such average results of atom per cent fission, F_T, obtained by the same analyst will approximate 0.8 per cent relative. Two such values should be considered suspect (95 per cent confidence level) if they differ by more than 1.9 per cent relative.

8.2 The multi-laboratory precisions for the average of duplicate determinations of F′, U′, and F_T are also given in Table I. The average difference between two such results obtained by different laboratories will approximate 1.7 per cent relative. Two F_T values should be considered suspect (95 per cent confidence level) if they differ by more than 4.2 per cent relative.

NOTE 1. - The precision estimates for F′ are based on an interlaboratory study on four samples including two samples of irradiated uranium fuel with 12.0 and 10.6 gigawatt days per tonne burnup, one calibration standard solution containing a mixture of Nd, U, and Pu equivalent to 13.1 gigawatt days

per tonne, and one solution of natural Nd. One analyst in each of 8 laboratories analyzed duplicate filaments prepared from each sample for a total of 58 reported determinations. Six determinations were unreported. No data were rejected. The precision estimates for U′ are based on an interlaboratory study on four samples including the same two irradiated uranium fuel solutions and two natural uranium solutions containing 2.50, 9.82, 10.77, and 12.50 mg U/ml. One analyst in each of three laboratories performed duplicate determinations and repeated one day later, for a total of 48 determinations. No data were rejected. The precision estimates for atom per cent fission are computed from the precision estimates of F′ and U′. Recommend Practice E 180 was used in developing these precision estimates. It should be noted that values in these studies were read from strip chart recorders. It has been reported (7) that the most important random error in isotopic analysis is due to the strip chart recorder, although recognition of this fact is not widespread.

TABLE I. - PRECISION OF ANALYSES

Value Measured	Single-Instrument Precision (1S) % relative	Multi-laboratory Precision (1S) % relative
F′	0.6	0.9
U′	0.4	1.2
F_T	0.7	1.5

9. ACCURACY (Bias or Systematic Error)

9.1 In mass spectrometry, the presence of a bias is possible, but mass spectrometers can be calibrated so that mass discrimination bias is eliminated. To accomplish this, measured mass ratios shall be bias corrected according to Section 5. It is expected that the method so calibrated will be free of bias and that the accuracy can be taken to be equal to the precision (see 8) except for some additional uncertainty in the fractional fission yield of Nd^{148}. The fractional yield for Nd^{148} in U^{235} thermal fission is reported to be 0.0166 ± 0.0003 [see Ref. (6)].

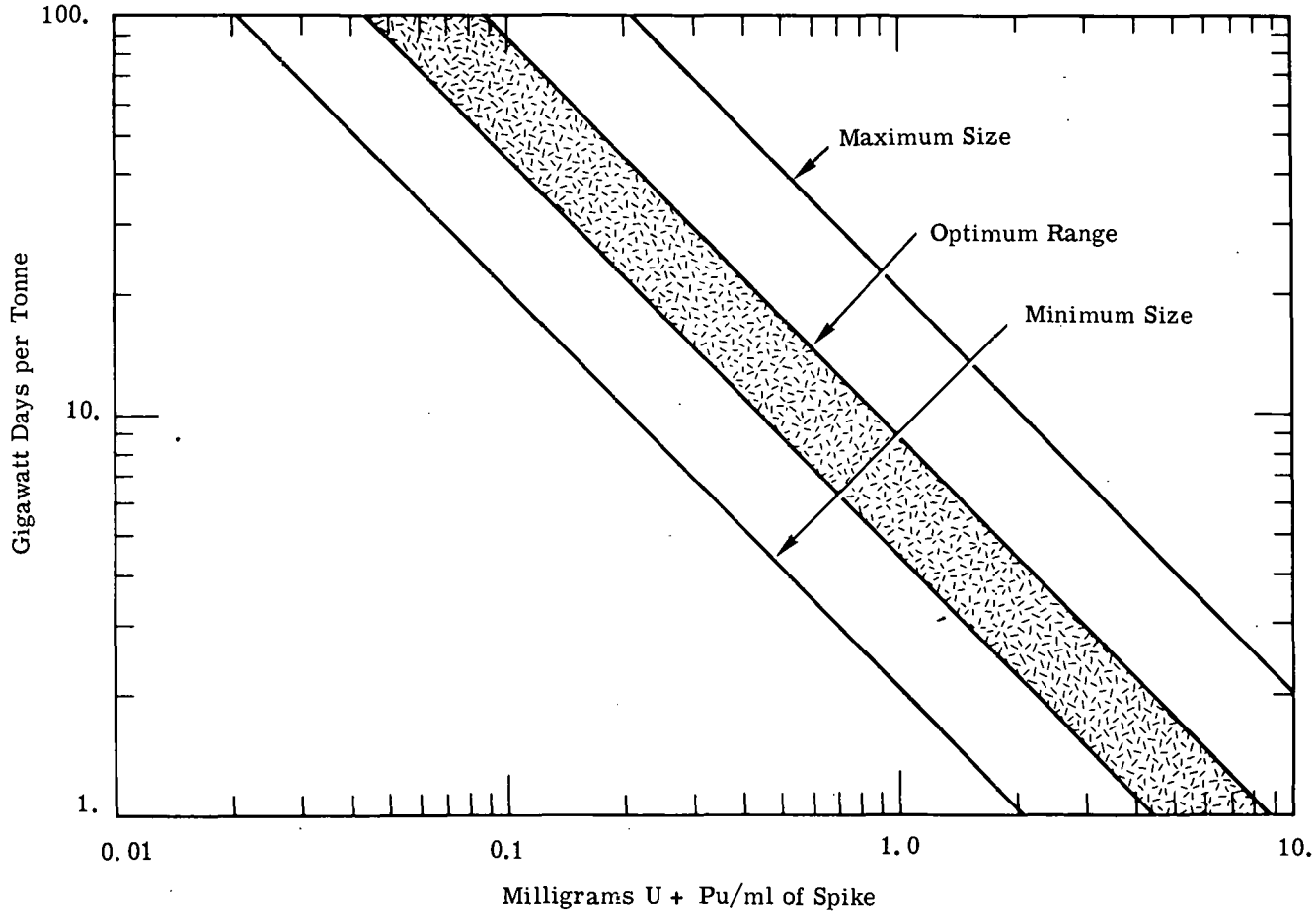

FIGURE A-1. Sample Size Required for Nd^{148} Analysis

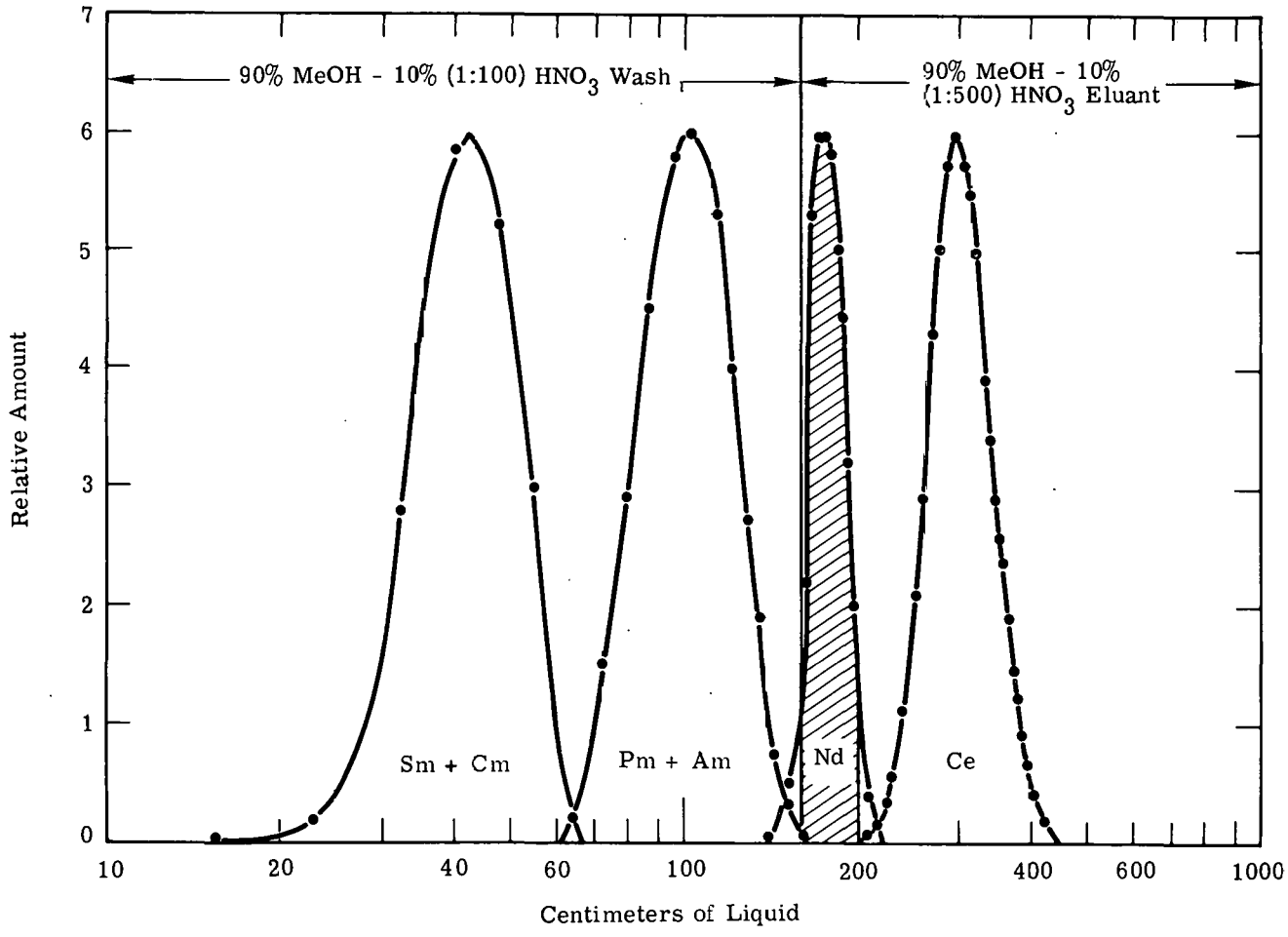

FIGURE A-2. Anion Exchange Chromatogram with Methanolic HNO_3 Eluant

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(2) B. F. Rider, J. P. Peterson, Jr., and C. P. Ruiz, "Determination of Neodymium-148 in Irradiated UO_2 as a Measurement of Burnup," Transactions of the American Nuclear Society, Vol. 7, No. 2, 1964, p. 350.

(3) B. F. Rider, C. P. Ruiz, and J. P. Peterson, Jr., "BURNUP: A FORTRAN IV Code for Computing Uranium and Plutonium Fuel Burnup from U, Pu, and Nd Mass Spectrometric Measurements," United States Atomic Energy Commission Doc., GEAP-5355, 1967.

(4) B. F. Rider, J. L. Jaech, J. P. Peterson, Jr., C. P. Ruiz, and F. R. Smith, "Accurate Nuclear Fuel Burnup Analysis XII," United States Atomic Energy Commission Doc., GEAP-4776, 1964.

(5) L. E. Weaver, P. O. Strom, P. A. Killeen, "Estimated Total Chain and Independent Fission Yields for Several Neutron-Induced Fission Process," United States Atomic Energy Commission Doc., USNRDL-TR-633, 1963.

(6) B. F. Rider, C. P. Ruiz, J. P. Peterson, Jr., and F. R. Smith, "A Survey and Evaluation of Thermal Fission Yields for U-234, Pu-239, U-233 and Pu-241," United States Atomic Energy Commission Doc. GEAP-5356, 1967.

(7) W. R. Shields, "Analytical Mass Spectrometry Section: Instrumentation and Procedures for Isotopic Analysis," NBS Technical Note 277, National Bureau of Standards, Washington, D. C., 1966, p. 16.

FOOTNOTES

[1]The numbers in parentheses and underscored refer to the list of references appended to this method.

[2]"Reagent Chemicals, American Chemical Society Specifications," Am. Chem. Soc., Washington, D. C. For suggestions on the testing of reagents not listed by the American Chemical Society, see "Reagent Chemicals and Standards," Joseph Rosin, D. Van Nostrand Co., Inc., New York, New York, and the "United States Pharmacopeia."

[3]1965 Book of ASTM Standards, Part 20.

[4]Dowex-1 resin (either Ag 1-× 2 or AG 1- × 4, 200-400 mesh) obtained from Bio-Rad Laboratories, 32nd St. and Griffin Ave., Richmond, Calif., has been found satisfactory.

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