140
10-20
No copy NTIS
No Stock (copy)

W. J. Gallagher
MASTER
BHO-67-4

EVALUATION OF ISOTOPIC SOURCE ASSAY SYSTEM
(ISAS)
UNDER COMMERCIAL OPERATING CONDITIONS

Final Technical Report

W. J. Gallagher

March 25, 1970 to March 25, 1971

Prepared for the U. S. Atomic Energy Commission
under Contract AT(30-1)-67
USAEC Plant Instrumentation Program

**UNITED
NUCLEAR
CORPORATION**

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights.

Printed in the United States of America

Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151

Price: Printed Copy: \$3.00; Microfiche: \$0.95

EVALUATION OF ISOTOPIC SOURCE ASSAY SYSTEM
(ISAS)
UNDER COMMERCIAL OPERATING CONDITIONS

Final Technical Report

W. J. Gallagher

March 25, 1970 to March 25, 1971

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Prepared for the U. S. Atomic Energy Commission
under Contract AT(30-1)-67
USAEC Plant Instrumentation Program

UNITED NUCLEAR CORPORATION
Commercial Products Division
365 Winchester Avenue
New Haven, Connecticut 06508

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

ABSTRACT

In cooperation with the U. S. Atomic Energy Commission, the United Nuclear Corporation, as part of the USAEC Plant Instrumentation Program, installed isotopic neutron source assay systems (ISAS) at two commercial facilities to determine the worth of the instruments for special nuclear materials control and safeguards under commercial operating conditions. The ISAS units were designed, fabricated, and installed by Gulf Radiation Technology (now Gulf Energy & Environmental Systems) under subcontract to United Nuclear Corporation.

The subcontract included consultation services of the Gulf team, Drs. T. Gozani, D. Costello, and R. Bramblett, during testing of the ISAS units and evaluation of the resultant data.

The ISAS units were used to measure a variety of compounds, mixtures, and alloys, as well as uranyl nitrate solution containing uranium of ^{235}U enrichments ranging from 2 to 97%. These materials were packaged within 5-in. diameter $\times$ 6 $\frac{3}{4}$ -in. steel cans, or 5 $\frac{3}{4}$ -in. diameter $\times$ 11-in. poly jugs of 1-gal volume. Up to 5 kg of ^{235}U per container was assayed without reaching the limit of ISAS. Determination of accuracy based upon three sets of comparisons between ISAS assays and chemical and mass spectrographic analytical data, obtained during reprocessing of the material, has indicated that $\pm 2\%$ accuracy can be obtained on dry homogeneous materials containing large quantities of uranium-235 when identical standards are used. The precision (reproducibility) of the assay system is, as is expected, controlled by counting statistics.

The sensitivity of the two ISAS units for unmoderated samples differs significantly, apparently due to the softer neutron spectrum in ISAS No. 1. ISAS No. 1 gives approximately twice the net signal for 300 grams of unmoderated ^{235}U as ISAS No. 2. ISAS No. 2 can detect 5 grams of ^{235}U distributed in 5 kg of Al_2O_3 in a one-gallon polyethylene jug. The lower limits of detection for ISAS No. 1, or for moderated fissile material in either unit, were not determined.

The units proved to be relatively trouble-free, the major operational failures occurring in the mechanical drive units for the sample carriers. The sample carriers of these two prototype ISAS units should be replaced with top-mounted, heavier sample carriers.

The cost of operation of the systems is relatively low and within the ranges which we have experienced for gamma counting.

The ISAS provides a means of quickly assaying the ^{235}U content of problem materials, such as bulk residues and slags, to a degree of accuracy which has until now been unattainable. There are many unanswered questions concerning ISAS (such as how well it can measure the fertile material content of enriched uranium bearing materials), but the system, as it presently exists, is an assay tool which will be valuable to special nuclear materials control and safeguards. Its utility could be substantially enhanced by improvement of the accuracy of measurement with small quantities of ^{235}U .

TABLE OF CONTENTS

1.	FINAL TECHNICAL REPORT	1
1.1	Introduction	1
1.2	Theory of ISAS Operation	2
1.3	ISAS Design	9
1.3.1	Source Container and Collimator (SC&C)	9
1.3.2	Fission Multiplicity Detector (FMD)	10
1.3.3	Control Console	12
1.4	ISAS Operating Techniques	13
1.5	Breakdown of Equipment	15
1.6	Measurement of ^{238}U	17
1.7	Calibration Curves	18
1.8	Precision (Reproducibility) of ISAS	20
1.9	Accuracy of ISAS	24
1.10	Design Differences vs Performance and Sensitivity	26
1.11	Time Required for Assay	30
1.12	Effect of Poly Sample Container Wall Thickness	30
2.	SUMMATION AND EVALUATION	33
2.1	Capabilities of ISAS	33
2.2	Evaluation of ISAS Usefulness to Special Nuclear Materials Control and Safeguards	33
	APPENDIX A - RADIATION DOSE RATES - ISAS NO. 2	35
	APPENDIX B - ISAS NO. 1 - CALIBRATION CURVES	37
	APPENDIX C - ISAS NO. 2 - CALIBRATION CURVES	47
	APPENDIX D - ISAS NO. 2 - SENSITIVITY TESTS	57
	APPENDIX E - MEASUREMENT OF ^{238}U	61

FIGURES

1.	General View of the ISAS (Isotopic Source Assay System)	3
2.	Fission Multiplicity Detector	4
3.	ISAS with Sample Container in Detector	5
4.	ISAS Vertical Scan Operation	7
5.	ISAS Fission Multiplicity Detector	11
6.	UO ₂ Oxide – ISAS No. 1	16
7.	Effect of Plastic Sample Container Wall Thickness on Signal – ISAS No. 2	31

1. FINAL TECHNICAL REPORT

1.1 INTRODUCTION

On March 25, 1970, Contract AT(30-1)-67 was awarded to the United Nuclear Corporation - Commercial Product Division (UNC-CPD), for the fabrication, testing, and evaluation of two isotopic source assay systems (ISAS), as part of the USAEC Plant Instrumentation Program. On April 27, 1970, United Nuclear Corporation subcontracted to Gulf General Atomic Incorporated (now Gulf Energy & Environmental Systems Company), the task of fabricating the two ISAS units and providing consultation during testing of the systems and evaluation of the resultant data.

The target accuracy for ^{235}U assay, relative to appropriate standards, was $\pm 2\%$ relative (at one standard deviation) for homogeneous samples containing greater than 300 grams of ^{235}U . Each ISAS unit was to be capable of separately measuring ^{238}U . Although the state-of-the art did not permit a quantitative estimation of the accuracy of the assay device for measurement of ^{238}U , the objective was to achieve $\pm 2\%$ relative accuracy when measuring the total uranium content of homogeneous samples containing greater than 300 grams of uranium.

The initial calibration of the ISAS units was performed by the Gulf Radiation Technology Division at the La Jolla, California, facility, using uranium standards prepared by United Nuclear Corporation. ISAS No. 1 was installed at the UNC-CPD Chemical Operations Plant, Hematite, Missouri, on October 26, 1970. ISAS No. 2

was installed at the UNC-CPD Recovery Operations Plant, Wood River Junction, Rhode Island, on November 14, 1970.

UNC-CPD personnel operated the two ISAS units to gain experience, and to prepare calibration curves for various types of enriched uranium-bearing material found in industry. In February 1971, the ISAS units were put into routine commercial use and assay data were collected for evaluation and comparison. In accordance with the concepts of the Plant Instrumentation Program, emphasis was directed toward strategic quantities of fissile material.

The following sections detail the findings and conclusions from this program.

1.2 THEORY OF ISAS OPERATION

A general view of the ISAS is shown in Fig. 1. It consists of two main parts: the high energy neutron isotopic source located in the Source Container and Collimator (SC & C) assembly, and the Fission Multiplicity Detector (FMD) and its associated electronics. (See Figs. 2 and 3.)

The FMD performs the function of detecting fission events. When a fission occurs, several neutrons (about 2.5 on the average) and gamma rays (about 7 on the average) are emitted simultaneously. Thus, a fission event is characterized by high multiplicity and can be distinguished from background events which are usually of much lower multiplicity or of uncorrelated single events. By requiring a coincidence between several detectors which are sensitive to gamma rays and neutrons, the fission events can be selected even in the presence of a relatively large background.

The FMD (see Figs. 2 and 3) is composed of four plastic scintillators. Two detectors are placed on either side of a sample cavity. Compton scattering cross-talk between the two adjacent detectors on a side is reduced by lead shielding. Since the plastic detectors have a large hydrogen content and are in close proxim-

Fig. 1 — General View of the ISAS (Isotopic Source Assay System)

Fig. 2 — Fission Multiplicity Detector

Fig. 3 — ISAS with Sample Container in Detector

ity to the sample cavity, a neutron shield (LiH or paraffin and elemental boron) is usually placed between the detectors and the sample to prevent neutrons moderated in the detector from returning to the sample cavity. This shield helps to reduce neutron self-shielding effects in the sample at the expense of a slightly reduced fission rate.

Fast electronics are used for processing the pulses from the detector. The anode pulses from the four photomultipliers are sent to four fast discriminators, where uniform pulses are generated which are then sent to various coincidence circuits. The chance coincidence contribution to the signal is small because of a narrow time gate (~ 60 nsec). The discriminators are set at a low level, which is just above the photo tube noise (approximately 100 keV γ -energy). Lead shielding (1/8-in. thick) surrounding the sample cavity is used to lower the count rate from low energy γ -rays which might originate in the sample. The lead shielding also reduces Compton cross-talk between detectors. The low bias improves the operation of the detector in two ways: (1) the detectors are more sensitive to neutrons from fission events, since, in general, the light pulses from protons (from neutron proton collisions) are much smaller than an electron with the same energy; and (2) by having a low bias, the detector efficiency is much less affected by gain shifts in the photo tubes (if the bias were at zero, the detector efficiency would be independent of the photo tube gain).

In order to approach uniform irradiation and response, the sample is rotated and vertically scanned. The scan is accomplished by a motor and gear assembly located at the bottom of the FMD. Fig. 4 illustrates the operation of the vertical scan, which is accomplished by moving the sample completely through the response region of the system. In Example A, it is assumed that all the fissile material is at the bottom of the container. The vertical scan is started with the fissile material completely out of the neutron beam. The count rate of the FMD, if the fissile material is far enough away, would just be the background. As the fissile material moves into the neutron beam, the count rate increases until the material

Fig. 4 — ISAS Vertical Scan Operation

reaches the center of the neutron beam where the count rate reaches a maximum. After the material passes through the center of the system, the count rate decreases again to the background level, when the fissile material is again out of the response region. In practice, a 500-sec traverse moves the sample through a total distance of 14.5 in. With a 0.6-kg ^{235}U sample (total height = 3 in.), the coincidence rate at both the upper and the lower limits of traverse is approximately 10% above background. The contribution of the sample to this coincidence rate is about 1.5% of the maximum coincidence rate. In Example B, the assumption is made that the same amount of fissile material as in Example A is concentrated at the top of the container. The response of the system to the material, of course, is the same, with the exception that it is shifted along the vertical axis the same distance as the difference in the placement of the material in Examples A and B. The same total counts will be obtained in both examples provided the vertical traverse is long enough so that the material in each case passes completely through the response of the system. From these two examples it is easily seen that only if the sample is scanned completely through the response of the system can the total count obtained from a given amount of material be independent of the vertical placement in the sample container.

Fertile material has a higher fission threshold than fissile material. The neutron energy required for fission (≈ 1 Mev) in ^{238}U is much higher than for ^{235}U , which fissions at thermal neutron energies. This difference in the fission properties of the two materials can be exploited to obtain the amount of both fertile as well as fissile material in a given sample. The ISAS has the built-in capability of measuring both types of material, by taking two separate measurements of the same sample; one measurement with an unmoderated neutron beam, and the second measurement with a moderated, lower energy neutron beam. The device has provisions for insertion of a moderator between the isotopic source and the sample.

1.3 ISAS DESIGN

ISAS consists of three physically separate components: a Source Container and Collimator, a Fission Multiplicity Detector, and a Control Console containing read-out equipment and sample motion (scan) controls.

1.3.1 Source Container and Collimator (SC&C)

The SC&C is a 24-in. diameter by 26-in. long stainless steel cylinder. All but a 6-in. diameter pipe through the center of the cylinder is filled with a neutron moderator (paraffin wax) mixed with thermal neutron absorbers (boric acid and lithium carbonate). The center pipe houses the isotopic source, the neutron collimator, and a movable shield plug.

The encapsulated source, within a small lead cylinder, is contained within the 1.5-in. diameter center hole of a larger 4-in. diameter by 6-in. long lead cylinder. This larger lead cylinder is positioned within the center pipe of the SC&C by means of the movable shielded plug.

The neutron collimator is positioned at the front of the larger lead cylinder containing the isotopic source. This collimator defines a 4-in. by 8-in. neutron beam at the center of the sample cavity in the ISAS Fission Multiplicity Detector. If needed, the collimator can be replaced with another of different size.

The sources used with the two ISAS units are ^{252}Cf . As of 7-6-70, the strength of these sources was:

<u>Source No.</u>	<u>Effective ^{252}Cf Content, μg</u>	<u>Neutron Emission Rate, n/sec</u>
118	$24.04 \pm 3.0\%$	$5.634 \times 10^7 \pm 3.0\%$
121	$23.77 \pm 3.0\%$	$5.572 \times 10^7 \pm 3.0\%$

1.3.2 Fission Multiplicity Detector (FMD)

The FMD is basically a lead-shielded aluminum cylinder, containing a 6-in. diameter center hole for accepting sample containers up to $5\frac{3}{4}$ -in. in diameter and 11-in. long. Within the FMD are two pairs of plastic scintillation detectors (Nuclear Enterprises Plastic Scintillation NE-102 Detectors). Each pair of detectors is mounted such that one detector is located directly over another detector and isolated from the other detector by a lead shield. The two pairs of detectors are located directly opposite each other, such that they "see" two sides of the sample. Each of the four detectors is optically coupled to a photomultiplier tube (see Fig. 5).

A neutron shield, within the FMD, provides low-energy neutron decoupling between the detectors and the sample (see Fig. 5). ISAS No. 1 utilizes a lithium hydride sample neutron shield, and ISAS No. 2 contains a paraffin shield loaded with 20% elemental boron.

The FMD detects a certain fraction of the number of fissions induced in the sample by the neutron beam. The basic detector efficiency for gamma rays has been calculated by Rad Tech to be 33% for 0.1 Mev energy, 21% for 1 Mev energy, and 8.3% for 10 Mev energy. This stated efficiency is equivalent to zero bias efficiency. The efficiency for detecting a fission event is 1.5% (three-out-of-four coincidence).

A sample drive mechanism, mounted beneath the FMD, rotates and raises/lowers the sample during neutron interrogation. Unless interrupted manually, the sample motion (and counting) stops automatically when the sample reaches either the upper or lower limit switch. The direction of vertical travel is automatically reversed when either limit switch is tripped.

During all operations covered in this report the ISAS units were adjusted to three-out-of-four coincidence, that is, the neutrons and/or gamma radiation

ISAS No. 1

A 16-in. diam
B 6-in. diam
C 6½-in.

ISAS No. 2

A 16¼-in. diam
B 6¼-in. diam
C 6½ in.

Scale: 1/8

Fig. 5 — ISAS Fission Multiplicity Detector

emitted from the sample being neutron interrogated had to be simultaneously detected by three out of the four plastic scintillation detectors before a count was recorded.

1.3.3 Control Console

The Control Console contains two separate systems, the sample motion (scan) controls, and the electronics for processing the pulses from the detectors.

A. Sample Motion Controls

This electromechanical system controls the rotation and vertical scanning of the sample within the Fission Multiplicity Detector. A single push-button activates both the sample motion and the counting system, or the counting system can be activated separately. A signal indicates when the sample carrier is in motion and in which vertical direction it is moving. The sample vertical speed is 1.75 in. per minute, while rotating at 28 rpm.

B. Pulse Processing Electronics

Each of the four photomultiplier tubes in the Fission Multiplicity Detector emits negative pulses from the anode and positive pulses from the 11th dynode. The Control Console has modular electronics to process and summarize these signals. The electronics consist of four fast channels to process the anode pulses, and one linear channel for the summation of the four dynode pulses.

The fast channels are made of one quad discriminator (LRS-321B) to bias out photomultiplier noise and to standardize the anode pulses. The rectangular output pulses from the discriminator are input to a coincidence circuit consisting of a logic mixer and another quad discriminator.

The pulses from the four dynodes are summed in the Linear Mixer and Delay and are transferred into the linear amplifier part of the spectrometer (Elscint

INS-11S), through a Linear Gate (EG&G LG 101/N) which opens only when the required coincidence has occurred.

Following the linear amplifier, the pulse height range of interest is defined by the window single channel analyzer (Elscont Integrated Nuclear Spectrometer INS-11S). Usually only the lower discriminator is used. The output of the single channel analyzer is recorded in a preset time/count scaler. The use of this single channel analyzer was intended to help distinguish neutron capture events of high multiplicity from the fission events, since, in general, much more energy is released from the fission event. It is not particularly effective in this application, and normal operation should be with the bias set very low to maximize the count rate.

1.4 ISAS OPERATING TECHNIQUES

To determine the ^{235}U content (or the total uranium content) of an unknown sample, the unknown is measured by ISAS for a predetermined length of time, and the normalized total net count is compared with a calibration curve drawn from data obtained by measuring known standard samples of the same type of material. Total U or ^{235}U is read directly from the calibration curve.

To calculate normalized total net count, three factors must be known or compensated for:

1. The ISAS measurement (gross count) of the unknown.
2. The shielding effects of the uranium and/or matrix material in the unknown, and the possible contribution to the signal caused by neutron interaction with material other than uranium. To compensate for, and/or reduce these effects, a background sample is measured for the same length of time as the unknown and the total count is subtracted from the total (gross) count of the unknown. The resultant number is the total net count of the unknown.

The background sample should be composed of material of composition, bulk, and density similar to the uranium-bearing material. Since the background coincidence rate is not sensitively dependent on the background sample material, e.g., 3 kg of Al_2O_3 or 13 kg PbO_2 increases the background by approximately 10%, the background measured with no sample has sometimes been used when large quantities of ^{235}U are being measured.

3. The "drift" of the ISAS unit. Drift is compensated for by repeated measurements of a normalization standard during each series of measurements of unknowns. Measurements of the unknown are normalized to this standard by dividing the total net count from the unknown by the total (gross) count from the standard. The resultant number is the normalized net count of the unknown.

During a series of measurements, the length of time for each measurement of the normalization standard is held constant, but this time may be different from the measurement time of the unknowns in the series.

Note: During measurements at UNC-CPD, the normalization standards were usually measured for 200 seconds, while the unknown samples were usually measured between 300 to 600 seconds depending on the sample count rate.

When the sample to be measured by ISAS has a relatively low count rate and/or contains relatively few grams of ^{235}U , thermalization of the neutron beam at the sample container (past the Fission Multiplicity Detector neutron shield) may be considered to increase the response from the ^{235}U . At present, because of

physical limitations, this technique is limited to containers of $5\frac{1}{4}$ -in. or less diameter. This technique was illustrated, using ISAS No. 1, by measurement of cans (5-in. diameter $\times$ $6\frac{3}{4}$ -in. long) of 3.20% enriched UO_2 for 622 seconds, with and without a $7/16$ -in. thick sleeve of paraffin around each can, as shown in the following tabulation and Fig. 6.

<u>Grams of Contained ^{235}U</u>	<u>Normalized Net Counts — without Paraffin Sleeve</u>	<u>Normalized Net Counts — with Paraffin Sleeve</u>	<u>Increase in Signal Due to Moderation</u>
5	0.32	0.34	4.6% Rel.
10	0.57	—	—
20	1.09	1.51	39.2
40	1.84	2.20	19.7
80	3.46	4.09	18.1
100	4.26	4.64	9.0

The effect is seen to be small and has not been exploited.

1.5 BREAKDOWN OF EQUIPMENT

The ISAS sample carrier consists basically of an aluminum cup on top of a motor-driven drive screw, mounted beneath the Fission Multiplicity Detector and extending up into it. Because of the long drive screw, it is necessary to drill a 28-in. deep hole (approximately 2 in. in diameter) in the floor, beneath the screw, or mount the unit on a 28-in. high platform. (Each UNC-CPD ISAS unit is positioned over a hole in the floor.)

When the sample carrier nears the upper vertical position limit, the aluminum cup on top of the drive screw touches the inner wall of the Fission Multiplicity Detector and vibrates. Approximately one to two man-hours of effort per week were required to re-align the sample carrier drive mechanism to prevent vibration and "chattering." This problem was substantially reduced on ISAS No. 2

Fig. 6 — UO_2 Oxide — ISAS No. 1 (3.2% Enriched U within 5-in. Diameter $\times$ 6 $\frac{3}{4}$ -in. Long Steel Cans; Spectrometer Baseline 0)

by fitting a thin Teflon sleeve around the exterior of the aluminum sample cup. The sample carrier drive screw is weakened by a deep key cut along its length and tends to bow under load. This drive screw was replaced in ISAS No. 2.

To eliminate vibration and binding of the sample carrier in ISAS No. 1, the operating personnel shortened the sample scan time, to prevent the sample carrier from reaching its lower vertical travel position where binding was occurring. Later investigation indicates this probably prevented the samples from receiving uniform irradiations. It appears likely that samples irradiated for the shorter time periods entered but did not travel through and out of the neutron interrogating beam. The effect of this is not quantitatively known.

A stronger, redesigned sample carrier should be fitted on top of the Fission Multiplicity Detector to eliminate vibration and the need for holes in the floor or platforms.

Late in the testing program, it was noted that the count rate from one of the photomultiplier tubes (Tube No. 1) in ISAS No. 2 fell off noticeably whenever the cable to this tube was moved. This was readily detected when measuring the normalization standard. The tube could be restored to normal operation by gently moving the cable. To avoid delaying the evaluation program, it was decided to continue operations and avoid disassembly of ISAS No. 2 until completion of the program. Later investigation revealed a broken connection in the cable base, resulting in an intermittent connection whenever the cable was moved. The broken connection undoubtedly hampered the reproducibility of ISAS No. 2 over an extended period of time.

1.6 MEASUREMENT OF ^{238}U

^{238}U has a fission threshold of approximately 1 Mev. Since the ^{252}Cf neutron source emits many neutrons above 1 Mev, fission events occur in the ^{238}U present

in the sample being irradiated. The relative responses per gram for ^{235}U and ^{238}U in highly enriched uranium has been obtained from ISAS-2 measurements with identically packaged 533-gram samples of 92.68% enriched and 0.053% depleted U_3O_8 . In these samples the $^{235}\text{U}/^{238}\text{U}$ response ratio was 5.3. Under the reasonable assumption that the ^{238}U response per gram is independent of uranium enrichment, and the less obviously valid assumptions that the ^{235}U response per gram is independent of enrichment over the high-enrichment range and that the ratio of response is insensitive to changes in the sample geometry, the above ratio has been used to correct ISAS data for enrichment differences between samples and calibration standards.

ISAS was designed to permit the measurement of the total uranium and the enrichment of low-enriched uranium by taking two measurements of the unknown sample. One measurement is made with an unmoderated neutron beam, and the second measurement is made with moderating material placed between the source and the sample to enhance the response from the ^{235}U . A cavity is provided in ISAS for insertion of moderating material.

Procedures for measurement of total uranium and enrichment are detailed in Appendix E. For the purpose of this report, an operational rather than a mathematical explanation is given.

This proposed technique has not been experimentally evaluated. For high-enrichment material where the enrichment is known, the additional effort is not believed to be warranted.

1.7 CALIBRATION CURVES

Enriched uranium-bearing materials commonly found within the commercial industry were standardized, packaged within 5-in. diameter by $6\frac{3}{4}$ -in. long steel cans or $5\frac{3}{4}$ -in. diameter by 11-in. long (1-gal volume) poly jugs, and mea-

sured by ISAS. Calibration curves were drawn from the resultant data. The following calibration curves can be found in Appendices B or C.

ISAS No. 1

<u>Standard Curve No.</u>	<u>Container</u>	<u>Material</u>	²³⁵ U <u>Enrichment</u>
1-1	Steel can	U ₃ O ₈ -Al ₂ O ₃ heterogeneous oxide	97.5%
2-1	Steel can	Impure ammonium diuranate	97.0%
3-1	Poly jug	Acid insoluble residue	92.6%
4-1	Steel can	UO ₂ oxide	3.2%
5-1	Steel can	UO ₂ pellets	2.01%
6-1	Steel can	Item product (Classified)	97.6%
7-1	Poly jug	UO ₂ oxide	97.6%
8-1	Steel can	Fluid bed reactor cleanout material	97.6%

ISAS No. 2

1-2	Steel can	U ₃ O ₈ -Al ₂ O ₃ heterogeneous oxide	97.5%
2-2	Steel can	Impure ammonium diuranate	97.0%
3-2	Poly jug	Acid insoluble residue	92.6%
4-2	Poly jug	Zirconia crucible pieces with U	97.0%
5-2	Poly jug	Assay tank solution	93.0%
6-2	Steel can	Item product (Classified)	97.6%
7-2	Steel can	Impure U ₃ O ₈	86.7%
8-2	Steel can	U ₃ O ₈ oxide final product	92.7%
9-2	Poly jug	Calcined ash (bomb slag)	86.7%

Due to sample neutron self-shielding, neutron multiplication, γ -ray attenuation, changes in the neutron flux by the matrix material, and count rate effects,

the calibration curves are not linear. Therefore, it is necessary to calibrate ISAS with a number of standard samples which completely cover the range of the expected assay.

Gulf Radiation Technology performed experiments to support the expectation that the pronounced nonlinearity at low-uranium content is principally a neutron self-shielding effect. If the nonlinearity were due primarily to gamma attenuation effects, then the deviation from a straight line would be greater for four-way coincidence than for coincidence between two detectors or the three-out-of-four coincidence. This was not found to be the case. Further, measurements were made by placing a small plate of ^{235}U metal in the sample cavity behind the sample such that the neutrons passed through the sample before striking the ^{235}U metal. The count rate from the ^{235}U plate was observed to decrease with increased loading of uranium in the sample in front of the plate. This also indicates neutron self-shielding.

Standard Curves #1-1 and #1-2 were based on measurements of U_3O_8 in a matrix of two to three kilograms of Al_2O_3 such that the total sample volume was preserved. These curves appear more nonlinear than those based on measurements of U_3O_8 alone, in which case the sample height varied. This may be a consequence of the variation of both the neutron self-shielding and the gamma-ray attenuation with uranium density.

1.8 PRECISION (REPRODUCIBILITY) OF ISAS

The precision of ISAS during a measurement series was determined by repeatedly measuring uranium standards and analyzing the standard deviation in the resultant series of data.

As expected, the rms deviation from the mean of a series of measurements is directly related to counting statistics. These precision measurements for ISAS-1 are generally consistent with the counting statistics. The 110-volt a-c

supply to ISAS No. 1 has had as much as 20% fluctuation, and this could contribute to the fluctuations at higher count rates.

The ISAS-2 data reported below show less fluctuation than would be expected on the basis of the number of counts involved. No significance has been attached to this observation.

ISAS No. 1 (All Measurements by the Same Operator)

<u>Gross Count Rate, counts/sec</u>	<u>Precision at One Standard Deviation</u>	<u>Number of Determinations</u>	<u>Measurement Time, sec</u>
3.3	±3.1% rel.	12 (empty detector)	200
10.4	1.7	4	500
15.1	1.2	4	500
27.3	1.2	8	500
59.2	1.1	95 (over 5 days)	200
173.2	0.7	39 (over 2 days)	200

Check measurements by a different operator, for a shorter time period, but otherwise under similar conditions, gave a comparable precision.

<u>Gross Count Rate, counts/sec</u>	<u>Precision at One Standard Deviation</u>	<u>Number of Determinations</u>	<u>Measurement Time, sec</u>
10.3	±1.3% rel.	10	300

ISAS No. 2

<u>Gross Count Rate, counts/sec</u>	<u>Precision at One Standard Deviation</u>	<u>Number of Determinations</u>	<u>Measurement Time, sec</u>
60.7	0.51	20	200
62.4	0.46	10	200
61.9	0.91	20	200

Abrupt changes in the temperature of the ISAS environment, such as a drop of approximately 20°F within 20 seconds, causes the count rate to rise approximately 5% relative. The assay system quickly stabilizes itself, within a few minutes, after the temperature becomes constant. An insulated sheet metal shield placed over the ends of the four photomultiplier tubes has significantly reduced the sensitivity of ISAS to abrupt temperature changes. Gradual temperature changes are no problem as they are compensated for by periodic measurements of the normalization standard.

The reproducibility of ISAS over an extended period of time has not been extensively evaluated due to lack of sufficient data obtained under identical operating conditions; however, the following suggests that 3 to 5% fluctuations in normalized response for well-characterized samples may be expected.

ISAS No. 1

3.20% enriched UO₂ oxide packaged within 5-in. × 6³/₄-in. steel cans:

<u>²³⁵U, grams</u>	<u>Normalized Net Signal</u>		<u>Nov. 10 Signal/ Nov. 12 Signal</u>
	<u>Nov. 10, 1970</u>	<u>Nov. 12, 1970</u>	
5	0.031 ± 0.003	0.035 ± 0.003	0.89
10	0.057 ± 0.003	0.060 ± 0.003	0.95
20	0.105 ± 0.003	0.103 ± 0.003	1.02
40	0.184 ± 0.004	0.182 ± 0.004	1.01

ISAS No. 1 (Continued)

^{235}U , grams	Normalized Net Signal		Nov. 10 Signal/ Nov. 12 Signal
	Nov. 10, 1970	Nov. 12, 1970	
80	0.334 ± 0.005	0.337 ± 0.005	0.99
100	0.419 ± 0.005	0.419 ± 0.005	1.00

All measurements agree within the statistical limits of error.

ISAS No. 2

93% enriched uranium – uranyl nitrate solutions, within $5\frac{3}{4}$ -in. $\times$ 11-in. poly jugs of one-gallon volume:

^{235}U , grams	Normalized Net Signal		Nov. 18 Signal/ Dec. 3 Signal
	Nov. 18, 1970	Dec. 3, 1970	
3.95	0.30 ± 0.01	0.29 ± 0.01	1.03
7.81	0.57 ± 0.01	0.58 ± 0.01	0.98
15.51	1.06 ± 0.01	1.11 ± 0.01	0.95
30.93	2.02 ± 0.01	2.10 ± 0.01	0.96
61.74	3.96 ± 0.02	3.73 ± 0.02	0.99
123.39	6.35 ± 0.02	6.25 ± 0.02	1.02
192.76	8.14 ± 0.03	8.26 ± 0.03	0.99

The only series of measurements which has been repeated over a substantial period of time is a subset of the U_3O_8 standards in ISAS-2. These data are plotted in Standard Curve #8-2 (Appendix C) and those measurements which have been repeated more than once are tabulated below.

92.68% Enriched U_3O_8 :

^{235}U , grams	Normalized Net Signal			
	12/10/70	1/26/71	3/8/71	5/6/71
98.9	0.190	0.214	0.188	0.194
296.6	0.498	0.517	0.500	0.489
494.3	0.793	0.922	0.815	—
790.9	1.153	1.26	1.16	—
988.6	1.50	1.67	1.53	—
1977.1	2.94	2.93	2.83	2.95

The data obtained on January 26 are erratic and perhaps should be ignored. If they are not, the rms deviation from the mean is 4.3%. If the January 26 data are deleted, the remainder of the data exhibits an rms deviation from the mean of 1.3%.

1.9 ACCURACY OF ISAS

The accuracy of ISAS, under specific conditions, is determined by comparing assay data obtained through measurement of certain lots of uranium-bearing material by ISAS, with assay data of the same lots of material obtained through analytical chemistry and mass spectrographic analysis.

A number of lots of enriched uranium-bearing scrap, of various chemical compositions, scheduled for recovery of the contained uranium at the UNC-Recovery Operations Plant, in Wood River Junction, Rhode Island, have been measured by ISAS No. 1 and/or ISAS No. 2. The following comparisons between ISAS assays and the results of chemical assay have been made.

1. Five 5-in. $\times$ 6 $\frac{3}{4}$ -in. steel cans of U_3O_8 final product, 97.1% enriched uranium, Lot SL-50, were measured by ISAS using a 600-sec traverse.

The U_3O_8 calibration data obtained with 92.68% enriched material were used and the data corrected for the difference in ^{238}U content as discussed in Section 1.6. The resulting ISAS determination of the ^{235}U content of these five cans was 14,520 grams. The uncertainty in this determination from counting statistics was 0.5%. Chemical and mass spectrographic analysis of the same material resulted in an assay of $14,796 \pm 90$ grams of ^{235}U .

2. Twelve 5-in. $\times$ $6\frac{3}{4}$ -in. steel cans of U_3O_8 oxide final product, 76.58% enriched uranium, Lot S-095, were measured by ISAS for 660 sec.

The 92.68% enriched calibration data for U_3O_8 were again used and corrected for enrichment. The ISAS assay for these 12 cans was 22,093 grams ^{235}U . In this instance the enrichment correction was 4%. The assay of the same material by chemical and mass spectrographic analysis was $21,188 \pm 51$ grams ^{235}U .

These two evaluations of clean, well-characterized material indicate differences of 1.9% and 4.3% between ISAS and chemical assay. The larger difference is associated with 76.58% enriched material, where a substantial correction was required to apply the 92.68% enriched calibration data. Calibration of ISAS with material of more than one enrichment is recommended.

3. Seventeen 5-in. diameter $\times$ $6\frac{3}{4}$ -in. long steel cans of Item Plant Scrap, 97.66% enriched uranium, Job H9011, were measured by ISAS for 600 sec.

The calibration data presented in standard curve #6-2 (Appendix C) were used to obtain ISAS assays. Five of these containers yielded net normalized responses which exceeded the range of the calibration curve. The curve was extrapolated. The ISAS assay totaled 39,225 grams ^{235}U . The chemical and mass spectrographic assay

totaled $35,885 \pm 90 \text{ g } ^{235}\text{U}$. Most of the 9% difference may be ascribed to an incorrect extrapolation of the calibration curve. However, the uranium density in these scrap materials is generally lower than in the clean calibration material, and part of the increased ISAS response should be associated with this reduction in uranium density.

1.10 DESIGN DIFFERENCES VS PERFORMANCE AND SENSITIVITY

The fission multiplicity detectors of ISAS Nos. 1 and 2 have small, but significant, design differences. ISAS No. 1 utilizes lithium hydride as the sample neutron shield (see Fig. 5), while ISAS No. 2 utilizes elemental boron plus paraffin for the corresponding part.

The sample neutron shield screens out thermal neutrons and, in effect, decouples the plastic scintillation detectors from the sample. Without this shield, thermal neutrons striking the detectors would form 2-Mev gamma rays and add to the total count rate; more significantly, neutrons thermalized in the detector could reach the sample. The lithium shield in ISAS No. 1 absorbs the thermal neutrons without emitting gamma radiation. The boron shield in ISAS No. 2 gives a harder neutron spectrum and forms 480-keV gamma radiation, which is shielded from the detectors by 1/8-in. lead.

Other, probably less significant, design differences are that the ISAS No. 2 sample cavity is 1/4-in. larger in diameter, and the distance between two opposite photomultiplier tubes is 1/4-in. larger than the corresponding dimensions in ISAS No. 1.

A comparison of assay data from ISAS No. 1 and ISAS No. 2 indicates that ISAS No. 1 is the more sensitive instrument for nonhydrogenous materials. This is illustrated by the following example.

Measurement of acid insoluble residue standards, 92.664% enriched uranium, packaged in 5³/₄-in. diameter × 11-in. long poly jugs.

<u>Jug No.</u>	<u>²³⁵U, grams</u>	<u>Net Signal-Total Counts</u>		<u>ISAS No. 1 Signal/ ISAS No. 2 Signal</u>
		<u>ISAS No. 1</u>	<u>ISAS No. 2</u>	
1	5.0	1,012	306	3.31
2	10.1	1,798	796	2.26
3	20.1	3,307	1,243	2.66
4	50.1	6,668	2,890	2.31
5	100.0	9,323	4,087	2.28
6	200.0	19,131	9,000	2.13
7	300.0	23,685	11,718	2.02

During these measurements all samples were interrogated for 500 seconds, with both ISAS No. 1 and ISAS No. 2 spectrometer baselines set at 175 and the upper windows at infinity.

This difference is attributed to a softer interrogating neutron spectrum in ISAS No. 1.

If the sample being assayed contains hydrogen, neutron moderation within the sample would be expected to decrease the difference between the two systems. This is illustrated by the following assay of ammonium diuranate containing a small (unknown) amount of water in addition to the chemically bound hydrogen.

The data for ISAS No. 1 were obtained at a baseline setting of 250 and have been corrected to correspond to a value of 175.

Measurement of impure ammonium diuranate standards, 97.008% enriched uranium, packaged in 5-in. diameter $\times$ 6 $\frac{3}{4}$ -in. long steel cans.

Can No.	²³⁵ U, grams	<u>Net Signal - Total Counts</u>		<u>ISAS No. 1 Signal/ ISAS No. 2 Signal</u>
		<u>ISAS No. 1</u>	<u>ISAS No. 2</u>	
3	10.04	828	347	2.4
4	20.07	1,505	794	1.9
5	50.18	3,720	2,520	1.5
6	100.4	8,600	7,170	1.2
7	200.7	18,700	17,119	1.09

The empty detector background with the baseline set at 175 is approximately 3.7 counts per second for ISAS No. 1 and approximately 4.7 counts per second for ISAS No. 2. This difference may be due to the two different sample neutron shields in these assay systems.

The differences in response of ISAS Nos. 1 and 2, which were initially complicated by differences in baseline settings, can be rationalized on the basis of a larger low-energy neutron component in the interrogating spectrum of ISAS No. 1. This rationalization is supported by noting that the 20% boron loaded paraffin surrounding the sample in ISAS No. 2 is expected to be more effective in suppressing the low-energy neutron component than the LiH in ISAS No. 1.

If the sensitivity of the ISAS system, s , is defined as that quantity of uranium whose net response in a standard traverse is twice the standard deviation in the background, and if it is assumed that the background coincidence rate has been very accurately determined, then

$$s = \frac{2 \sqrt{\text{background counts}}}{\text{Counts/g of uranium}}$$

In the case of ISAS No. 2 for highly enriched nonhydrogenous material, such as U_3O_8 , where the background for a 600-second traverse is approximately 3000 coincidences and the counts per gram are approximately 23, the lower limit of detection, s , is 4.7 g of ^{235}U . Substantially smaller quantities can be detected in aqueous solution. The limit of detection can be reduced by reducing the background or increasing the coincidences per gram of uranium. Both avenues should be exploited.

The counts per gram of uranium can be increased by:

1. Increasing the interrogating neutron flux;
2. Moderating the interrogating neutrons;
3. Eliminating the vertical scan.

The neutron flux can be increased by increasing the source strength and by removing the portion of the sample shield between the source and the sample. Increasing the source strength under present conditions is not expected to be effective, since the uncertainty in the background is not dependent solely upon counting statistics and is believed to be limited to approximately 2% of the background. Removal of a portion of the sample shield may be effective and should be tried.

Moderating the neutrons will increase the ^{235}U fission rate, but only at the expense of larger self-shielding corrections.

Eliminating the vertical scan is feasible for physically small samples. For large samples such as one (1) gallon bottles of residues in which the uranium distribution is unknown, this approach can not be used.

Reduction of the background coincidence rate will require modification of the physical construction of the ISAS units and should be investigated.

1.11 TIME REQUIRED FOR ASSAY

The cost of an assay by ISAS is directly related to the length of time required to complete a measurement of an unknown sample and convert the measurement to grams.

Under the assumption that measurement of a single sample requires a 200-sec determination with the normalization standard at the beginning and at the end of the measurement, and that 8-minute scans of both the sample and an appropriate background sample are required, the time required for a single sample assay is 25 minutes.

With multiple samples of the same type, a single background sample determination is normally adequate and the number of normalization standard measurements per sample is reduced so that the average time per sample is reduced to 15 minutes for a batch of six (6) similar samples.

1.12 EFFECT OF POLY SAMPLE CONTAINER WALL THICKNESS

The wall thickness of one-gallon poly jugs, used for storage of uranium bearing materials, varies according to the manufacturer. The poly jugs used by United Nuclear Corporation have a wall thickness of approximately 0.043 in. The polyethylene acts as a moderator and enhances the ^{235}U signal.

A series of measurements (see Fig. 7) in which samples containing approximately 250 g ^{235}U contained in a 1 $\frac{3}{4}$ -in. diameter glass bottle were wrapped with varying thicknesses of polyethylene indicated that an increase of 3 to 5% in the coincidence rate could be obtained within the range of normally available polyethylene bottle wall thicknesses.

Therefore, it is desirable to standardize containers for both unknown and calibration samples.

Fig. 7 — Effect of Plastic Sample Container Wall Thickness on Signal — ISAS No. 2 (Measured for 200 sec; Spectrometer Baseline 175)

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

2. SUMMATION AND EVALUATION

2.1 CAPABILITIES OF ISAS

ISAS can assay large quantities of ^{235}U in the form of dry oxides, alloys, or mixtures contained in dry nonhydrogenous materials, up to one gallon in volume, for which standard reference samples are available, to an accuracy which is of the order of 2%.

A few grams of ^{235}U can be detected but not accurately assayed in a dry sample containing up to several kg of inert material. Up to 5 kg of ^{235}U per container has been measured without reaching the limits of the system.

In the assay of aqueous solutions of uranium compounds, where the hydrogen content can be accurately represented in standard samples, the sensitivity of the measurement is increased by a factor of at least 20, and the accuracy of the measurement is comparable to that obtained with large hydrogenous samples.

Errors will be introduced if the hydrogen content of the samples to be assayed differs significantly from the hydrogen content of the standards used to establish the respective calibration curve.

2.2 EVALUATION OF ISAS USEFULNESS TO SPECIAL NUCLEAR MATERIALS CONTROL AND SAFEGUARDS

ISAS should prove to be one of the most valuable nondestructive assay tools available for control and safeguard of uranium. It is capable of routinely nonde-

structively assaying the ^{235}U contents of bulk, dense, uranium-bearing materials to a useful degree of accuracy. It can be operated by a technician. The cost of operation is relatively low, so that the maximum number of measurements may be taken, and tighter limits of error may be established for special nuclear material inventories.

ISAS provides a means of quickly measuring such problem materials as heterogeneous bulk scrap, residues, calcined ash, and slag, to a degree of accuracy which has not routinely been achieved by other assay methods. Measurement of such problem materials will help to resolve a Materials Control/Safeguards problem which has plagued industry and resulted in significant shipper/receiver differences.

While the system has been found to be quite useful in its present state, its utility would be substantially enhanced by:

1. Improving the reliability and flexibility of the sample traversing mechanism.
2. Reduction of the time required to assay a sample.
3. A substantial decrease in the limit of detection for dry samples so that reliable (10%) assay data can be obtained with a few grams of ^{235}U .
4. Incorporation of a device to indicate, and, if possible, quantitatively define the hydrogen content of a sample.

APPENDIX A - RADIATION DOSE RATES - ISAS NO. 2

A survey of neutron and gamma dose rates in the area around the Isotopic Source Assay System was made using a PNR-4 and the NC-2650. The ISAS sample chamber and moderator chamber were empty during these measurements. The results are tabulated according to the numbered locations on Fig. A.1.

<u>Location</u>	<u>Dose Rate, mrem/hr</u>		
	<u>Neutron</u>	<u>Gamma</u>	<u>Total</u>
1	4.0	0	4.0
2	2.5	0	2.5
3	1.75	0	1.75
4	20.0	0.55	20.6
5	3.0	1.0	4.0
6	3.0	0.55	3.55
7	0.75	0.25	1.0
8	2.0	0.05	2.05
9	1.2	0.02	1.22
10	0.4	0.05	0.45
11	0.3	0.1	0.4
12	0.75	0.1	0.85
13	0.3	0.08	0.38
14	0.1	0.05	0.15
15	0.1	0.05	0.15
16	4.5	1.4	5.9
17	4.0	1.2	5.2
18	3.5	1.6	5.1
19	3.0	1.7	4.7
20	4.0	1.5	5.5
21	2.5	0.55	3.05
22	0.2	0.08	0.28
23	1.0	0.18	1.18

Fig. A.1 — Isotopic Source Assay System No. 2

APPENDIX B – ISAS NO. 1 – CALIBRATION CURVES

Fig. B.1 — Standard Curve #1-1 — $\text{U}_3\text{O}_8\text{-Al}_2\text{O}_3$ Heterogeneous Oxide — ISAS No. 1
(97.5% Enriched U; Matrix of Approximately 3 kg Al_2O_3 ; Packaged within 5-in. $\times$
6 $\frac{3}{4}$ -in. Steel Cans; Measured for 300 sec; Spectrometer Baseline 250)

Fig. B.2 — Standard Curve #2-1 — Impure Ammonium Diuranate — ISAS No. 1 (97.008% Enriched U; Packaged with 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; Contains Fe, Cr, Ni, and Possibly Small Amount of Water; Measured for 500 sec; Spectrometer Baseline 250)

Fig. B.3 — Standard Curve #3-1 — Acid Insoluble Residue — ISAS No. 1
(92.6% Enriched U; Standards Are within 5³/₄-in. × 11-in. Poly Jugs;
Unknown H₂ Content; Matrix Material is Sand, Al(OH)₃, ZrO₂, and Filter
Aid, Totaling Approximately 3.6 kg; Measured for 500 sec; Spectrom-
eter Baseline 175)

Fig. B.4 — Standard Curve #4-1 — UO_2 Oxide — ISAS No. 1 (3.20% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; 500-sec Measurements; Spectrometer Baseline 250)

Fig. B.5 — Standard Curve #5-1 — UO_2 Pellets — ISAS No. 1
(2.01% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in Steel Cans;
Spectrometer Baseline 225)

Fig. B.6 — Standard Curve #6-1 — Item Product — ISAS No. 1 (97.6% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; Baseline: 225; Background Standard was 0.4% Depleted U of Similar Composition; 622-sec Counting Time; Normalization Standard Was a U Disc)

Fig. B.7 — Standard Curve #7-1 — UO_2 Oxide — ISAS No. 1 (97.64% Enriched U; Packaged within $5\frac{3}{4}$ -in. $\times$ 11-in. Poly Jugs; Measured for 300 sec; Spectrometer Baseline 250)

Fig. B.8 — Standard Curve #8-1 — Fluid Bed Reactor, Cleanout Material —
ISAS No. 1 (97% Enriched Item Product in NbOCl_3 Matrix; Packaged within
5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; Measured for 500 sec; Spectrometer Baseline 250)

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

APPENDIX C - ISAS NO. 2¹ - CALIBRATION CURVES

Fig. C.1 — Standard Curve #1-2 - Heterogeneous Oxide - ISAS No. 2
(97.5% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; Matrix
of Approximately 3 kg Al_2O_3 ; Measured for 605 sec; Spectrometer
Baseline 175)

Fig. C.2 — Standard Curve #2-2 — Impure Ammonium Diuranate — ISAS No. 2 (97.008% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; Contains Fe, Cr, Ni, and Possibly Small Amount of Water; Measured for 610 sec; Spectrometer Baseline 175)

Fig. C.3 — Standard Curve #3-2 — Acid Insoluble Residue — ISAS No. 2 (92.6% Enriched U; Standards Are within 5³/₄-in. × 11-in. Poly Jugs; Unknown N₂ Content; Matrix Material is Sand, Al(OH)₃, ZrO₂, and Filter Aid, Totaling Approximately 3.6 kg; Measured for 510 sec; Spectrometer Baseline 175)

Fig. C.4 — Standard Curve #4-2 — Zirconia Crucible Pieces with U — ISAS No. 2 (97.008% Enriched U Packaged within 5³/₈-in. × 11-in. Poly Jugs Containing Approximately 4.5 kg Zirconia; Measured for 600 sec; Spectrometer Baseline 175)

Fig. C.5 — Standard Curve #5-2 — Assay Tank Solution Standards — ISAS No. 2 (93% Enriched Uranyl Nitrate Solution; Sample Volume 1-gal Plastic Jugs; Spectrometer Baseline 175)

Fig. C.6 — Standard Curve #6-2 — Item Product — ISAS No. 2 (97.6% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in Steel Cans; 500-sec Measurement Time; Spectrometer Baseline 175)

Fig. C.7 — Standard Curve #7-2 — Impure U₃O₈ — ISAS No. 2
(Contains Fe, Cr, Ni within 5-in. × 6³/₄-in. Steel Cans; 86.71%
Enriched U; Measured for 610 sec; Spectrometer Baseline 175)

Fig. C.8 — Standard Curve #8-2 — U_3O_8 Oxide Final Product, Homogeneous — ISAS No. 2 (92.68% Enriched U; Packaged within 5-in. $\times$ 6 $\frac{3}{4}$ -in. Steel Cans; 610-sec Measurement Time; Spectrometer Baseline 175)

Fig. C.9 — Standard Curve #9-2 — Calcined Ash (Bomb Slag) with U — ISAS No. 2
(86.71% Enriched U; Packaged within 5 $\frac{3}{4}$ -in. $\times$ 11-in. Poly Jugs; Measured for
500 sec; Spectrometer Baseline 175)

APPENDIX D - ISAS NO. 2 - SENSITIVITY TESTS

Fig. D.1 — 97% Enriched U — ISAS No. 2 (Item Product; Dry Heterogeneous Mixture with 5000 g Al_2O_3 within 1-gal Plastic Jug, $5\frac{3}{4}$ -in. Diameter $\times$ 11-in. Long; Measured for 500 sec; Spectrometer Baseline 175)

3/17/71

ISAS No. 2

SENSITIVITY MEASUREMENTS OF
DRY HETEROGENEOUS MATERIAL

Instrument Settings

Baseline = 175, Gain Multiplier = 4

Upper Window = 0, H.V. = 1550

Notes:

1. Background standard is 500 g Al_2O_3 within a $5\frac{3}{4}$ -in. diameter $\times$ 11-in. long plastic jug.
2. Normalization standard is a poly moderated U-Al plate, containing 116 g U, 108 g ^{235}U .
3. Matrix material is 5000 g Al_2O_3 within a $5\frac{3}{4}$ -in. diameter $\times$ 11-in. long plastic jug. During this test, uranium (97% enriched Item Product) was added to this matrix material in increments of 1 g ^{235}U each. After each addition of ^{235}U , the matrix material was shaken vigorously, by hand, 20 times.

TABLE D.1 — ISAS NO. 2 — SENSITIVITY MEASUREMENTS OF DRY HETEROGENEOUS MATERIAL
(3/17/71)

<u>Sample</u>	<u>Time, seconds</u>	<u>Scan Position</u>	<u>Gross Counts</u>	<u>Net Counts</u> $\pm$	<u>Statistical Error</u>	<u>Normalized (to ^{235}U) Net Signal</u>
Normalization standard	200	Static	13,502	—	—	—
Normalization standard	200	Static	13,731	—	—	—
Empty detector chamber	495	Down	2,435	—	49	—
Background standard	497	Up	2,833	—	53	—
Background standard	497	Down	2,696	—	52	—
Normalization standard	200	Static	12,927	—	—	—
Matrix material + 1 g ^{235}U	499	Up	2,596	—	51	—
Background standard	497	Down	2,720	—	52	—
Normalization standard	200	Static	13,040	—	—	—
Matrix material + 2 g ^{235}U	498	Up	2,769	49	74	0.004 ± 0.006
Normalization standard	200	Static	13,478	—	—	—
(Interrupted measurements approximately 1/2 hour)						
Normalization standard	200	Static	13,015	—	—	—
Matrix material + 3 g ^{235}U	499	Up	2,760	120	74	0.009 ± 0.006
Background standard	498	Down	2,640	—	51	—
Normalization standard	200	Static	12,987	—	—	—
Matrix material + 4 g ^{235}U	499	Up	2,757	117	73	0.009 ± 0.006
Matrix material + 5 g ^{235}U	498	Down	2,840	200	74	0.015 ± 0.006
Matrix material + 6 g ^{235}U	499	Up	2,840	257	74	0.020 ± 0.006
Matrix material + 7 g ^{235}U	498	Down	2,890	307	74	0.024 ± 0.006
Normalization standard	200	Static	12,926	—	—	—
Background standard	499	Up	2,583	—	51	—
Background standard	498	Down	2,584	—	51	—
Matrix material + 8 g ^{235}U	500	Up	2,822	238	74	0.019 ± 0.006
Matrix material + 9 g ^{235}U	498	Down	2,918	334	74	0.026 ± 0.006
Normalization standard	200	Static	12,804	—	—	—
Matrix material + 10 g ^{235}U	500	Up	2,906	304	75	0.024 ± 0.006
Background standard	498	Down	2,602	—	51	—
Normalization standard	200	Static	12,655	—	—	—

APPENDIX E – MEASUREMENT OF ^{238}U

USE OF THE ISOTOPIC SOURCE ASSAY SYSTEM (ISAS) FOR THE DETERMINATION OF ^{238}U AND USE WITH LOW-ENRICHMENT MATERIALS

D. G. Costello

A. Use of the ISAS for Determining the Amount of Uranium in Unknown Samples for Low-Enrichment Materials when the Enrichment is Known

If a neutron has an energy greater than the fission threshold for ^{238}U (≈ 1 Mev) it can cause a fission event to take place in the ^{238}U as well as in ^{235}U which may be present. The ^{252}Cf neutron source emits some neutrons above this energy, and thus causes fission events to occur in ^{238}U which is placed in the sample cavity of the ISAS. The ISAS therefore will work with ^{238}U in the same way as for ^{235}U . The only difference is that the system's response to an amount of ^{238}U is smaller than the response to the same amount of ^{235}U .

For high enrichments, the response of the system to a given amount of uranium is mostly due to the ^{235}U present, since in comparison the ^{238}U response is very small. For low enrichments, the ^{238}U response does become important since it would be a larger part of the total response. For an enrichment of approximately 3%, the response of ^{238}U will equal the ^{235}U response.

If the enrichment of low-enrichment material is known, then the calibration of the ISAS for assay work follows the same procedure as for high-enrichment

materials. That is, a set of standards is prepared of the particular enrichment to be measured. These standards need to be of the same chemical form and in a matrix material which is similar to the material of the unknowns. After suitable standards have been made and a calibration curve obtained, the amount of uranium in an unknown sample may be obtained in the same manner as for high-enrichment material.

B. Method for Determining the Total Amount of Uranium as Well as the Enrichment for Use with Low-Enrichment Material

To employ this method, several sets of standards of different enrichments are necessary. It is imperative, of course, that the uranium be in the same chemical form in each of these sets, as well as in the same matrix material. After suitable standards have been obtained, the ISAS can be calibrated for determining not only the total amount of uranium in an unknown sample but also the enrichment of the material.

Two calibration curves are obtained for each of the different enrichment standards. One set of calibration curves is obtained by operating the ISAS in the usual manner, that is, with no moderating material in the neutron path between the ^{252}Cf source and the sample. Fig. E.1 shows a set of calibration curves that might be obtained for 0% (depleted uranium), 1%, 1.5%, 2%, 3%, 4%, and 5% enrichments. The curves in Fig. E.1 as well as Fig. E.2 are for illustration only, and do not represent experimental measurements. A second set of measurements is obtained from each of the standards by placing a moderating material in the tray which is located between the sample cavity and the neutron source. This moderating material will tend to slow down the neutrons before they interact with the sample, and this will enhance the ^{235}U response while at the same time decreasing the systems response to ^{238}U . Fig. E.2 illustrates the results which might be obtained with a moderated neutron beam for the same set of standards.

Fig. E.1 — The Yield vs Mass with No Moderation for Various Enrichments

Fig. E.2 — The Yield vs Mass with Moderation for Various Enrichments

The method for obtaining the assay from these two calibration curves can best be explained by the following two examples.

Example 1:

Suppose it is desired to determine the uranium content and also the enrichment of an unknown sample (it is assumed, of course, that this unknown is of the same chemical form and in the same matrix material as the standards from which the two sets of calibration curves were obtained). The unknown is run through the ISAS twice, once without a moderator and the second time with the moderator present. Now suppose 0.6 (the ratio of the sample counts to the calibration standard) is obtained for the measurement without the moderator, and 0.4 is obtained with the moderator in the neutron beam. Dashed lines have been drawn in Figs. E.1 and E.2 which illustrate these measurements. Now if the enrichment is not known, the amount of uranium could not be obtained from either of the measurements taken singly. For instance, referring to Fig. E.1, if the enrichment was 5%, then the amount of uranium would be 0.3 kg; if the enrichment was 4%, then the amount would be 0.43 kg, etc. Table E.1 shows the amount of uranium which would be present for the various enrichments for the two cases: with moderator and without moderator.

TABLE E.1 — EXAMPLE 1

<u>With Moderator</u>		<u>With No Moderator</u>	
<u>Enrichment</u>	<u>Mass, kg</u>	<u>Enrichment</u>	<u>Mass, kg</u>
5%	0.07	5%	0.3
4%	0.18	4%	0.43
3%	0.27	3%	0.66
2%	0.43	2%	1.0
1.5%	0.93	1.5%	1.2
1%	1.7	1%	1.35
		0%	1.50

Total uranium mass = 1.22 kg.
Enrichment = 1.4%.

If the enrichment vs the mass is plotted for both cases, we obtain two curves as shown in Fig. E.3. Notice that the two curves cross at a point corresponding to an enrichment of 1.4% and a mass of 1.22 kg. Now, since the true enrichment and the true mass of the unknown sample is the same in the two cases, this point of intersection of the two curves then represents the enrichment and the mass of the unknown amount of uranium.

Example 2:

The same procedure is followed in this example as for Example 1. Here the yield with no moderator is 0.64 and with a moderator the yield is 0.76. Again, the mass for the various enrichments is obtained for the two cases, and the enrichments are listed in Table E.2. These values are plotted in Fig. E.4, and the point of intersection of the two curves represents the enrichment and the mass of the unknown sample. For this example the enrichment is found to be 4% and the mass is 0.48 kg.

TABLE E.2 — EXAMPLE 2

<u>With Moderator</u>		<u>With No Moderator</u>	
<u>Enrichment</u>	<u>Mass, kg</u>	<u>Enrichment</u>	<u>Mass, kg</u>
5%	0.25	5%	0.34
4%	0.48	4%	0.48
3%	0.72	3%	0.66
2%	1.80	2%	1.09
1.5%	—	1.5%	1.30
1%	—	1%	1.44
0%	—	0%	1.61

Total uranium mass = 0.48 kg.
Enrichment = 4%.

Fig. E.3 — The Enrichment vs Mass for Yields of 0.6 without Moderation and 0.4 with Moderation

Fig. E.4 — The Enrichment vs Mass for Yields of 0.64 without Moderation and 0.76 with Moderation