Conf-710637--31

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**ACTIVE ASSAY OF FISSIONABLE MATERIALS
AT THE LOS ALAMOS
NONDESTRUCTIVE ASSAY AND STANDARDS LABORATORY***

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**Paper presented at the
Twelfth Annual Meeting
of the**

**INSTITUTE OF NUCLEAR MATERIALS MANAGEMENT
West Palm Beach, Florida
June 29-July 1, 1971**

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At LASL the development and application of active nondestructive assay technique has been and continues to be a major effort in the program of safeguards research. Emphasis so far has been on using neutrons, produced by either a machine or by radioactive sources, as the interrogating radiation. Howard Menlove, in a companion paper, has already discussed the application of radioactive neutron sources to the nondestructive assay of fissile material. Most technique and signature development has been done using the Van de Graaff accelerator and 14 MeV neutron generator installations at LASL. Practical field experience has been obtained with the MONAL which, since the last INMM meeting, has been at National Lead Company of Ohio and most recently completed a tour at Nuclear Materials and Equipment Corporation. Several hundreds of samples have now been analyzed using active techniques. Time permits only the discussion of a few examples to illustrate the progress in selected areas.

The first example is the receiver verification of 320 kg of highly enriched uranium (93% ^{235}U) in the form of UAl_x powder packaged in 90 containers (~ 2 kg U each). The UAl_x powder is quite uniform so that chemical sampling of each of the cans could provide a straightforward verification of

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the shipment. However the packages were sealed in an inert atmosphere so that opening, for the purpose of obtaining a sample, and resealing the packages plus the processing of 90 chemical samples would be both time consuming and expensive. A nondestructive assay of such a shipment is highly desirable. Because of the high density of this material passive assay is very difficult. The normal method of neutron interrogation, delayed neutron detection was chosen. Figure 1 shows the irradiation geometry employed. A measurement on an individual can required about three minutes. The measurements were done for three separate shipments of about thirty cans each. The assay of the third and last shipment (28 cans) was completed in one day, literally on a "while you wait" basis, that is the truck carrying the samples arrived in the morning and departed later the same day with the entire load of cans assayed nondestructively. All assays were performed relative to one of the cans chosen as a reference standard. Because of differences in packaging and amount of fissile material involved, the assay results are reported in three groups. The first group had a package which consisted of a steel inner and outer container; the second group had a polyethylene bottle as the inner container; the third group consisted of one package with a steel inner and outer container which had only about a quarter of the uranium content of the standard. The results for the three groups are shown in Table I. Multiple sampling and chemical assay of the material in one can of each of the three groups was done to establish the accuracy as well as the bias of the complete 90 can inventory. A comparison of the results of the sampling procedure is shown in Table II. The effect of the polyethylene bottle in group two was not significant. The overall agreement between all three parties in the assay is very good. It must be stated that the accuracy obtained is largely due to the uniform contents and the controlled geometry of the samples. Nonetheless when these conditions are met, this experiment shows that nondestructive assay by neutron interrogation can provide quite accurate and rapid verification of received material.

The second example is quite different from the first involving the assay of 144 inventory verification samples containing only subgram to gram quantities of fissile material. The samples were all uranium and ranged from highly enriched to only a few percent enrichment. It was necessary that all measurements be completed within two weeks. The samples were then sent to the New Brunswick Laboratory where the entire sample was completely dissolved and chemically assayed. The highly enriched samples were measured with three separate techniques; Van de Graaff subthreshold neutron interrogation, ^{235}U content; 14 MeV neutron interrogation, total uranium; passive gamma counting, ^{235}U content. Most of the low enrichment samples were done using only the Van de Graaff and the 14 MeV neutron generator. A few of the samples, some obviously wet, were compounds containing hydrogen. Many of the samples were measured twice by all techniques. In all, over 500 separate measurements were made in two weeks.

At present the data are in the process of being properly statistically analyzed. However a plot, in the form of a bar graph which shows the frequency of occurrence of a given percentage difference between the non-destructive and chemical results, provides a convenient method of comparison. The first graph, Fig. 2, represents data obtained, for each of the three techniques, from approximately sixty samples of highly enriched material. The data show a negative bias of between two and three percent, all of the nondestructive assays predicting less material than was found by chemical analysis. The reasons for this bias have not yet been traced. The width of the curves is consistent with the precision that would be predicted from the counting statistics of the measurements. The second set of graphs (Fig. 3) represents thirty-nine samples of low enrichment oxide powder. The Van de Graaff data shows a wide spread due to the lack of counting statistics. This situation can be readily improved by providing more sample. Most of these samples contained less than 50 mg of ^{235}U . The graph of the measurements of total uranium using 14 MeV neutron

interrogation has a width consistent with the counting statistics but now exhibiting a positive bias with respect to the chemical assays, this time predicting more material. The third set of graphs (Fig. 4) is obtained from measurements on thirty-three pellets. Van de Graaff data for ^{235}U show little bias while the 14 MeV neutron measurement of total uranium again indicates a negative bias between 2 and 3%. The number of data points corresponding to the samples containing hydrogen was too few to be represented by graphs. The Van de Graaff measurements showed a maximum of 10% increase in response due to the hydrogen content. Because of the harder interrogating spectrum, the 14 MeV neutron assay for total uranium is little affected by the hydrogen content. Results of the 14 MeV neutron interrogation and passive gamma ray counting of these samples were similar to those shown in Figs. 2 (passive) and 3 (Accelerator 1). For the assay of the 144 samples six standards were used. The reference fissile material in the standards was carefully evaluated both with respect to enrichment and total uranium content by the LASL analytical chemistry group (CMB-1).

Besides the important matter of indicating the existence of bias, the measurement of this large a number and variety of samples yielded information in other areas. It has already been demonstrated that ^{235}U and ^{238}U can be measured separately utilizing only the 14 MeV neutron generator plus appropriate spectrum tailoring. However the combination of the high sensitivity (about 10 mg ^{235}U) Van de Graaff subthreshold neutron interrogation for ^{235}U with 14 MeV neutron interrogation for total uranium proved to be very efficient and versatile. For a single assay method a normal work-day output of twenty to thirty samples was achieved. Including data reduction, sample handling and monitoring, etc., a given assay method may require the efforts of two to three people. With automated in-line data processing and sample handling, the output per man-hour can probably be more than doubled. Most of the work with small samples has been done using a small vial as the primary container. This container although very convenient for obtaining and handling the samples does not provide the best

geometry for nondestructive assay. A thin pancake-shaped container would afford the opportunity for better fission chamber coupling to the sample in the case of neutron assay and simplify attenuation corrections for gamma counting. The development of a packaging system which retains the simplicity of the vial yet more closely meets the requirements for nondestructive assay offers the potential for substantial improvement in the accuracy of the measurements. Practical implementation of these high throughput assay techniques as a complement and supplement to traditional chemical methods can go a long way toward keeping up with the requirements for inventory verification of a growing nuclear industry.

As was mentioned earlier the MONAI has just completed a tour at Nuclear Materials and Equipment Corporation*. At NUMEC a total of 314 bottles of uranium scrap in three major categories was measured by a combination of six nondestructive assay techniques:

1. The active can counter with ^{235}U and ^{238}U fission chamber monitoring capable of determining ^{235}U content and, by the two-spectra neutron interrogation method, capable of determining ^{238}U content as well,
2. The Ge(Li) gamma scan system,
3. A 5 x 5 in. NaI crystal with sample rotation set up to count the 1001 keV line from the decay of ^{238}U ,
4. A portable NaI detector used as an enrichment meter and air filter counter,
5. A neutron coincidence counter,
6. The neutron coincidence counter in conjunction with a Pu-Li source used for a preliminary test of the random source technique for determining ^{235}U content.

*Gratitude is expressed to J. E. Lovett and the personnel at NUMEC for the cooperation and courtesy extended to LASL personnel during the MONAI visit to NUMEC.

A description of how each of these techniques was employed as well as a summary and comparison of the results is presented in LA-4705-MS.

The first of these six techniques, the active can counter provides another example of the application of active nondestructive assay. Figure 5 is a diagram of the original active can counter. To satisfy the requirements for isotopic assay of uranium, the single plutonium fission chamber is replaced with a ^{235}U chamber and a ^{238}U fission chamber. Also a mechanism is provided to move a tungsten-lead block in front of the target. The spectrum shift with and without the block is sufficient to provide the two spectrum irradiation necessary for isotopic assay. (1)

A particular category for which the can counter was used for assay was miscellaneous scrap. This category includes such items as vacuum sweeper cleanout, air filter media, air filter shakeout, grinder grit, and incinerator ash. This category is characterized by: low uranium concentration (90% of the bottles had less than 100 grams ^{235}U), varying enrichments (10-90%), wide range of matrices, significant amounts of hydrogen, and often gross inhomogeneities within the can. Ninety-four of these samples were suitable for both assay in the can counter and passive assay by counting the ^{235}U gamma ray using Ge(Li). A comparison of the results obtained from these two techniques for the total amount of ^{235}U in these samples yielded agreement to within 3%. In general the experience at NUMEC showed that the relatively rapid measurement rate on entire containers enables the nondestructive techniques to provide more complete, detailed information on a larger number of samples and thus keep tighter control on individual categories. Because the entire container is measured, the vexing problems of representative sampling are bypassed. The two techniques of gamma scanning and neutron interrogation both overlap and complement one another. Gamma scanning is not influenced by hydrogen but is density limited at high U concentrations. Neutron interrogation while sensitive to hydrogen, gives accurate assay results over a wide range of U loadings or in other high-Z materials. In both systems an indication

of their limitations is provided by either an attenuation measurement or a fission chamber ratio. The MONAL capability for independent verification of results is extremely valuable for the evaluation of assay techniques since frequently no other assay values are available for comparisons.

Recently LASL has acquired a set of BWR fuel rods (Dresden type; 53 in. long) loaded with full density fuel pellets at 2.34% ^{235}U enrichment. A series of measurements using the Van de Graaff to produce subthreshold neutrons were made on these rods in the standard 6 x 6 BWR fuel element array as shown in Fig. 6. Provision has been made for vertical translation and rotation. There is also a steel reflector which is used to flatten the spatial response. This reflector has been removed to show the bare BWR element. The line of small detectors in front of the delayed neutron slab detector are ^4He prompt neutron detectors. The results to date are sufficiently encouraging to predict that it is feasible to design a neutron interrogation system utilizing a Van de Graaff to measure the total ^{235}U content of cold whole power reactor fuel elements to within 1-2%. As with any 1% measurement care must be taken to minimize background variations, spatial sensitivities, electronic drifts, etc. Using commercially available fuel element handling equipment, assay time for an entire element would be ~ 20 minutes.

The examples discussed above, chosen to illustrate progress in quite different application areas, represent only a part of the research effort in active assay. It is appropriate before closing to mention some of the other topics. Calculations are being and have been performed⁽²⁾ to provide information on how to better compensate for the presence of matrix materials as well as the effects of varying fissile material density. Calculations are being used to investigate the application of active assay to irradiated fuels. A calculational method^(3, 4) has been developed for the lead slowing-down spectrometer which takes into account all nuclear interactions as well as the effects of different spatial modes. Counting rates calculated using this method agree well with available experimental values. Small

sample assay of fissile compounds which are difficult to analyze chemically, e.g., Th-U-C beads, is being investigated. A study of sampling/chemical assay vs. nondestructive assay is being carried out for uranium-bearing organic ash. The data so far indicate that even when care is taken to mill, blend and sample the ash, a representative sample is difficult to obtain. On occasion, gross discrepancies have been observed.

Our experience involving a large number of samples differing widely in geometry, matrix materials and fissile content has shown that neutron interrogation is indeed a broadly applicable tool for nuclear material assay. The number of options available, i.e., radioactive source, machine source, delayed and/or prompt neutron detection, etc., provide a basis on which to design systems to meet a rather wide spectrum of requirements both economic and technical.

References

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3. C. R. Weisbin, R. A. Forster, D. B. Smith and N. Corngold, "Space and Time-Dependent Slowing Down in Heavy Media," Trans. Am. Nucl. Soc. 14, 386 (1971).
4. C. R. Weisbin, R. A. Forster and D. B. Smith, LA-4705-MS, pp. 22-24 (1971).

List of Figure Captions

- Figure 1.** Set-up at the LASL Nondestructive Assay and Standards Laboratory for neutron interrogation and delayed neutron assay of uranium aluminide samples. A UAl_x can to be assayed is shown inside a thermal neutron shield can (top removed) which is positioned between the neutron interrogation source on the left and the high-efficiency delayed neutron detector on the right (scale in inches).
- Figure 2.** Comparison of small sample assay results, high enrichment material.
- Figure 3.** Comparison of small sample assay results, low enrichment material.
- Figure 4.** Comparison of small sample assay results, pellets.
- Figure 5.** Active interrogation system for can assay.
- Figure 6.** Experimental arrangement for assay of power reactor fuel elements at the A-1 Van de Graaff accelerator. The Van de Graaff target is on the right and the ^3He delayed neutron slab detector, together with the bare ^4He prompt neutron detectors are on the left.

TABLE I

SUMMARY OF LASL NONDESTRUCTIVE ASSAY RESULTS

<u>Group</u>	<u>Total g U-235 NLCo</u>	<u>Total g U-235 LASL</u>	<u>Total g U-235 Difference^a</u>	<u>Fractional Difference^b</u>	<u>Mean^c</u>	<u>Standard Deviation^d</u>
1	183,512	184,245	+ 733	0.40%	1.0037	0.95%
2	113,885	113,812	- 73	0.06%	0.999	0.92%
3	817	751	- 66	8.1%	---	---
--	298,214	298,808	594	0.2%	---	---

^a (Total g U-235 NLCo) - (Total g U-235 LASL)

^b ((Total g U-235 Difference)/(Total g U-235 NLCo)) x 100

^c The mean of (Total g U-235 LASL)/(Total g U-235 NLCo) for each can

^d The standard deviation of (c).

TABLE II

COMPARISON OF CHEMICAL ANALYSIS AND
NONDESTRUCTIVE ASSAY RESULTS ON
THIRTEEN CANS OF HIGHLY-ENRICHED
URANIUM ALUMINIDE

<u>UAl Lot and Container No.</u>		<u>NLC_o. Inventory g ²³⁵U</u>	<u>ICPP Chem. g ²³⁵U</u>	<u>LASL NDA, g ²³⁵U*</u>
9836-2	4	3236.42	3231	3231
9837-5	6	4540.63	4534	4572
9836-2	3	3217.57	3208	3228
9837-5	5-3	3391.54	3380	3425
Composite #1		4286.00	4287	4252
Composite #2		4540.00	4541	4468
9835-60-3	7	2226.26	2219	2204
9834-60-6	3	2503.06	2500	2498
9834-60-5	6	3437.65	3445	3489
9835-60-4	5	3705.51	3724	3712
9834-60-1	5	3799.56	3791	3806
9835-60-1	2	801.02	801	750
9835-60-1	6	3206.39	3216	3192
TOTALS		42,893.43	42,877	42,827

* ICPP's chemical assayed content of Lot No. 9836-2, Container 4, used as standard.

Figure 1

HIGH ENRICHMENT SAMPLES

Figure 2

LOW ENRICHMENT SAMPLES

NUMBER OF MEASUREMENTS

Figure 3

PELLETS

NUMBER OF MEASUREMENTS

PERCENT

Figure 4

Figure 5

Figure 6