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THE NUCLEAR SAFEGUARDS RESEARCH AND DEVELOPMENT PROGRAM of the LOS ALAMOS SCIENTIFIC LABORATORY

A Program Review Covering Calendar Year 1968

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Introduction: Background and Scope of Program

The Los Alamos Scientific Laboratory has undertaken an extensive research and development program in the technology of inspection, accountability, and surveillance of fissionable material and fission devices to meet the growing urgent need for more effective national and international control of strategic nuclear materials. The implementation of an effective nuclear safeguards and materials management system requires direct physical methods of detecting, identifying, and quantitatively analyzing fissionable materials in various practical configurations containing both fissionable and nonfissionable materials. To be most effective and useful, such assay methods should be nondestructive, rapid, accurate, and capable of being carried out under a wide range of both laboratory and field conditions; e. g., in situ or on-line assay in materials processing plants, or in mobile isotopic assay stations for in-the-field use. The difficulties of representative sampling in chemical (destructive) assay of heterogeneous and complex systems are largely obviated in new nondestructive assay methods, mainly because of their characteristic high penetrability through bulk materials.

Nondestructive assay methods can be divided into two main categories:

(1) passive assay, and (2) active interrogation. Passive assay methods involve observation of both neutrons (from spontaneous fission or [α, n] reactions) and gamma rays (emitted following α decay) which are uniquely characteristic of individual fission species. The naturally occurring gamma lines having sufficient intensity for passive assay applications are typically a few hundred kilovolts or less in energy and hence have limited penetrability through dense materials. Also, in the case of highly radioactive samples (e.g., "spent" reactor fuel elements) any passive gamma-ray "signatures" are completely masked by the high radiation level of the sample. Thus for many practical assay problems, such factors as lack of penetrability, inherently high radioactivity levels, etc. severely limit the usefulness of passive methods, and one must employ active interrogation techniques. It should also be noted that, from an inspection and surveillance standpoint, active interrogation techniques are inherently more difficult to subvert or circumvent than are the simpler passive techniques.

Active interrogation involves the use of an external source of neutrons or photons to induce fissions in the material under investigation. Neutron sources were chosen for active interrogation in the LASL safeguards program because of (1) the high effective penetrability of fast neutrons in nuclear materials generally; (2) sharp, well-defined neutron fission thresholds which provide incisive isotopic discrimination; (3) readily

available, simple, inexpensive, compact neutron sources (e.g., D-D, D-T neutron generators, ^{252}Cf, etc.) of the required intensity and reliability for practical assay applications.

In active interrogation methods, quantitative assay is based on detailed observations of the delayed and prompt neutrons and gamma rays from fission. The delayed regime has the advantages of complete time-separation from the interrogating pulse, and permits the use of simple, inexpensive counting circuitry and data reduction equipment. Emphasis at LASL is placed on the development of very high efficiency detectors so as to maximize sensitivity and signal-to-noise characteristics while minimizing the interrogating-neutron-source strength with its associated shielding requirements.

The characteristic differences in yields and kinetic response of the delayed neutrons from the various fission species provide a unique method for analysis of individual isotopes in unknown mixtures of fissionable and nonfissionable materials. The experimental techniques involved in delayed neutron assay are rapid, nondestructive, and comparatively simple and inexpensive. For relative isotopic assay, no absolute calibrations (of source, detectors, fission rates, etc.) are required, and all measurements are well separated in time from the interrogating neutron pulse as well as time-dependent perturbations from prompt neutrons such as higher-mode decay, diffusion and thermalization effects.

Delayed fission gamma rays and prompt neutrons and gamma rays may be used either in combination with delayed neutrons, or separately, to provide characteristic signatures of individual fission species.

New nondestructive assay techniques developed on the basis of these signatures are being intercompared with the conventional chemical assay methods in use for many years in the various pilot-plant operations at LASL which represent essentially every stage of industrial fabrication, recovery, and processing of nuclear materials. Full exploitation of the various active and passive nondestructive assay methods under development at LASL promises practical solutions to a wide range of safeguards and nuclear materials accountability problems throughout the nuclear industry.

LASL Program Objectives

The purpose of the LASL program is to explore and develop practical new approaches to the nondestructive detection, identification, and quantitative assay of fissionable materials, as well as to improve on assay methods used in the past. These approaches include detailed investigations of the kinetics, yield and energy characteristics of delayed fission neutrons, prompt fission neutrons and gamma rays, and fission-product delayed gamma rays. Such basic data, which provide the fundamental signatures for various fissionable materials, are being applied in the development of

complete systems for nondestructive isotopic assay, and the early application of such systems to practical safeguards and nuclear materials management and accountability problems in the expanding nuclear industry, as well as to U. S. and international NPT-inspection requirements.

These broad objectives are being pursued via several parallel efforts at LASL:

1. Measurements of fundamental fission signatures whenever existing data are inadequate for assay applications. The required basic measurements include: absolute delayed neutron yield versus energy of the neutron inducing fission; group abundances and decay constants of delayed neutrons for 14-MeV neutron induced fission; time-dependent energy spectra of delayed neutrons; delayed gamma-ray lines characteristic of individual fission species which have suitable intensity and energy for assay applications.
2. Development of practical assay methods through experimental measurements and parallel theoretical calculations of delayed neutron and gamma-ray response of various practical configurations (e. g., fuels and fuel components, small samples and scrap) to pulsed neutron interrogation.
3. Development and adaptation of detectors, instrumentation, data acquisition/reduction equipment, etc. required for both

passive and active interrogation in safeguards R&D.

4. Development and adaptation of compact, intense neutron sources, source moderation and shielding, hot-cell manipulating facilities, sample handling equipment, etc. required for neutron interrogation and assay methods.
5. Direct intercomparison of newly developed nondestructive assay methods and results for typical reactor fuels, small test samples, and scrap produced at LASL and other nuclear facilities, with conventional destructive chemical assay as carried out at the AEC New Brunswick Laboratory, at LASL, and elsewhere.
6. Adaptation and integration of proven new assay methods into in-plant and mobile assay systems (e. g., roadable van) suitable for practical field use by the AEC and subsequently the nuclear industry.

Progress to Date in LASL R&D Program

Progress to date in each of the above-mentioned categories of work at Los Alamos can be summarized as follows:

1. Fundamental Data, Measurements, and Analyses

The fundamental data required for the practical application of delayed

neutron assay techniques include absolute delayed neutron yields (per fission) as a function of incident neutron energy. Absolute yield measurements or delayed-to-prompt neutron yield ratios provide a basis for effective discrimination between the thermally-fissioning species (notably ^{239}Pu and ^{235}U). Recent measurements[1] of the absolute yields of delayed neutrons from 3.1 and 14.9 MeV neutron-induced fission have been carried out for all of the major fission species (^{233}U, ^{235}U, ^{238}U, ^{239}Pu, and ^{232}Th). These measurements, summarized in Table I, show clearly that the delayed neutron yield per fission of every isotope studied decreases significantly in going from 3.1 MeV to 14.9 MeV fission--a result which is expected from the known behavior of fission mass and charge distribution but which is in direct contrast to previous measurements at other laboratories in both the U. S. A. and the U. S. S. R.

TABLE I

MEASURED ABSOLUTE DELAYED NEUTRON YIELDS

(Delayed Neutrons/Fission)

Indicated uncertainties are standard deviations. All yield values have been corrected to 100% isotopic purity.

Isotope	14.9 MeV Fission Yield	3.1 MeV Fission Yield	Yield Ratio: 3.1 MeV / 14.9 MeV
^{239}Pu	.0043 ± .0004	.0069 ± .0007	1.60 ± .09
^{233}U	.0043 ± .0004	.0077 ± .0008	1.79 ± .10
^{235}U	.0095 ± .0008	.018 ± .002	1.89 ± .11
^{238}U	.0286 ± .0025	.049 ± .005	1.71 ± .10
^{232}Th	.031 ± .003	.060 ± .006	1.94 ± .11

Basic delayed fission gamma-ray data may provide unique isotopic signatures or complement the delayed neutron assay methods already developed, depending on the specific gamma-ray characteristics measured. The gross delayed gamma-ray intensity (total photons per fission-sec) over the early time range from ~ 0.1 sec to 100 sec after fission depends on the isotope undergoing fission in much the same way as the delayed neutron emission. The combined measurement of both delayed neutron and gamma-ray response to active interrogation should prove useful for assay applications in which the environment of the nuclear material is unknown; i.e., because of large differences in the attenuation characteristics of neutrons and gamma rays, such combined data can give important information about the materials surrounding, or interspersed with, the fissionable material under investigation.

The yields of individual early delayed gamma-ray lines can be used for the assay of fissionable materials using techniques already developed for routine activation analysis. This method consists of detecting characteristic high-energy gamma rays from fission products by means of a high resolution Ge(Li) system in the time region from ~ 1 sec to 1 hour after exposure of the sample to a neutron beam. Of particular interest for active interrogation would be expected large differences in the yields of high-energy (penetrating) gamma rays from $^{235}U(n, F)$ and $^{239}Pu(n, F)$. In this case interrogation with subthreshold neutrons could be used to

assay ^{239}Pu and ^{235}U separately, in the presence of a large quantity of ^{238}U, as occurs in power reactor fuels. Examples of preliminary fission delayed gamma-ray spectra measured at LASL for ^{235}U(n, F) are shown in Fig. 1. As additional fundamental high resolution gamma-ray data are obtained in the course of this basic measurement program, the merits of gamma-line signatures for fissile material assay can be more fully evaluated.

The characteristic resonance structure in the neutron fission cross sections of the different fissile isotopes offers additional methods for nondestructive assay. This structure is most pronounced in the neutron energy range from 0.3 eV to roughly 10 keV where the resonance peaks are typically orders of magnitude larger than the valleys. These characteristics have been applied at LASL in development of the "resonance self-indication" technique for nondestructive assay.[2] This technique employs a steady-state beam of epithermal neutrons which is passed through a sample of fissile material and thence to a series of thin-foil fission detectors containing the same fissile isotope(s) as the sample being assayed. The responses of these detectors are sensitive to the resonance absorption lines in the transmitted flux corresponding to the resonance reaction peaks. This method is especially sensitive for ^{239}Pu assay because of the large (3000 barn) resonance in ^{239}Pu at 0.3 eV.

Another direct result of resonance self-indication studies at LASL

is the experimental delineation of the effectiveness of ^{239}Pu, ^{235}U, and ^{233}U neutron beam filters for enhancing fissile isotope discrimination (notably, to provide increased discrimination between ^{235}U and ^{239}Pu). The use of beam filters can also be combined with other characteristic signatures such as hard gamma lines from neutron-induced fission and neutron capture gamma rays. In this connection it is noteworthy that a suitable combination of assay techniques and signatures may often be used to provide an extremely effective "hybrid" method, specially tailored for a particular assay application.

2. Development of Practical Nondestructive Assay Methods

Some of the neutron interrogation techniques which are being developed at LASL for nondestructive assay are described briefly here, and representative assay results for a wide range of practical fissionable material configurations are given.

Delayed neutron assay techniques are based on two major approaches--the so-called "delayed neutron yield" and "kinetic response" methods. The yield method of assay is quite similar to the repetitive pulsing technique employed in the recent absolute delayed neutron yield measurements at LASL.[1] The unknown samples are repetitively irradiated with short pulses (50-100 ms) alternating with delayed neutron counting intervals of similar duration. For this mode of irradiation, measured delayed neutron

counting rate is essentially proportional to time-integrated delayed neutron yields, and in turn to the amounts of the fissionable materials present. Thus using a suitable flux monitor and/or fission monitor, the absolute amount of material present can be determined from measured delayed neutron yield as compared to the yield from a standard of known mass. This assay technique has been applied to MTR-type fuel elements and mockups with an assay accuracy of better than 1% over a wide range of fuel loadings; representative assay data for both bare and heavily-shielded elements are tabulated later in this section.

The kinetic response method of determining relative isotopic abundance consists basically of irradiating an unknown sample with fast neutrons and observing the delayed neutron response as a function of time. Delayed neutron decay is characterized by six exponential decay periods, τ_i, each having characteristic group abundances, a_i, which are significantly different for the various fission species.* The differences in abundances of the six delayed neutron groups result in readily measurable differences in the shape of delayed neutron decay following neutron interrogation. These decay-shape differences have been formulated in

*For example, the abundances of the longest (55 sec) delayed neutron group are nearly equal for ^{235}U and ^{238}U, but the abundance of the shortest delayed group ($\sim$ 0.2 sec) is a factor of seven greater in ^{238}U than in ^{235}U. Detailed delayed neutron group characteristics for all the major fission species are contained in Ref. 3, Chapter 4.

terms of experimentally measurable functions, $R_{f\pm}$ and $S_{f+/\Delta}$, and corresponding isotope discrimination ratios.[4] The relative isotopic abundance of a series of composite systems made up of two-inch diameter discs have been determined by analyzing the measured composite-system response curve as a linear superposition of normalized pure-isotope response curves. At the present stage of development, the kinetic response method is capable of determining relative isotopic abundances typically to 2-3% accuracy.

In both the delayed neutron yield and kinetic response methods just described, the energy of the interrogating neutrons can also be varied to take full advantage of the greatly increased isotopic discrimination afforded by the subthreshold and superthreshold fission characteristics of the various isotopes present in composite systems. In particular the use of subthreshold and superthreshold neutron interrogation provides a straightforward separation of the response of the fissile isotopes (e. g., ^{233}U, ^{235}U, ^{239}Pu) and the fertile isotopes (e. g., ^{238}U, ^{232}Th). Variability of neutron energy over the range relevant for threshold discrimination has been achieved for 14-MeV neutron sources by placing moderating, or "spectrum tailoring", assemblies around the source. These techniques are described in detail in section 4, under the topic of neutron sources and spectrum tailoring.

In addition to experimental measurements, both neutron transport

and Monte Carlo calculational techniques are being used to investigate the effects of neutron multiplication on observed delayed neutron response from systems of various size, geometrical configuration, and composition. The calculations provide invaluable guides for the development of practical assay systems, e. g., quantitative methods for scaling or interpolating between standard reference samples are being developed by means of Monte Carlo calculations.

Most practical applications of nondestructive assay methods can be divided into three major categories:

A. Reactor fuels (e. g., fabricated fuel rods, plates, discs, pins, feed materials, molten salt fuels);

B. Small test samples, e. g., fissionable material standards, prototype and experimental materials, compounds, mixtures, process-line samples in various physical and chemical forms;

C. Scrap, e. g., from isotope separation, fuel reprocessing and fuel fabrication plants.

Progress in each of these categories is summarized below·

A. Fuel assay results include assay of MTR elements, both bare and in lead radiation shields (see Figs. 2 and 3). In the case of bare fuel elements, delayed neutron assay for total amount of ^{235}U has been demonstrated to be accurate to ~ 0.4%, using appropriate standards.[5] Some typical experimental assay data for bare MTR fuel elements are shown in Table II.

TABLE II

DELAYED NEUTRON ASSAY OF ABSOLUTE AMOUNTS OF FISSILE MATERIAL IN MTR-TYPE FUEL ELEMENTS[5]

14-MeV (D-T) Interrogating Neutrons

Actual Weighed ^{235}U Content (grams)	Delayed Neutron Assay Determination (grams)
339.95 (fully loaded)	Calibration Point
320.95	320.9
302.15	303.2
283.95	283.5
265.45	266.5
247.05	249.9
	Average Deviation: 0.4%

Further, when the fuel element is enclosed in a massive lead shield (of thickness comparable to that of spent-fuel-element shielding casks), there is little perturbing influence on the results and accuracy of fuel element assay using neutron interrogation and the delayed neutron response method of assay. Typical assay results for heavily shielded MTR fuel elements are presented in Table III. The obvious practical implications for assay and burnup determinations of spent fuel elements in lead shielding casks are currently being investigated in the laboratory.

TABLE III

DELAYED NEUTRON ASSAY OF ABSOLUTE AMOUNTS OF FISSIONABLE MATERIAL IN HEAVILY SHIELDED MTR-TYPE FUEL ELEMENTS[6]

(Fuel Element Completely Enclosed in 4-inch Thick Lead Shield)

Actual Weighed ^{235}U Content in Element (grams)	Delayed Neutron Assay Determination (grams)	Deviation
340.0 (fully loaded)	Calibration Point	----
321.0	322.2	- 0.37%
302.3	304.8	- 0.82%
282.6	286.2	- 1.28%
	Average Deviation:	0.82%

LASL is developing the delayed neutron assay technique for nondestructive on-line isotopic assay of ^{233}U-Th fuel-salt mixtures for the Oak Ridge molten salt breeder reactor, MSBR. Actual MSBR fuel is a mixture of BeF_2, LiF, ThF_4 and $^{233}UF_4$, normally molten in the operating MSBR reactor, but solidifying into a highly heterogeneous marble-like solid (cf. Fig. 4) at room temperature. It has proved extremely difficult, if not practically impossible, to obtain representative samples for conventional wet chemistry assay techniques--hence the necessity for a nondestructive assay capability. Using LASL-prepared standards, MSBR

fuel samples were analyzed for absolute ^{232}Th content to well within 1% accuracy as determined by intercomparison with ORNL wet chemistry analyses.[7] Typical assay results are presented in Table IV. In addition, passive assay for thorium based on detection of ^{232}Th high-energy gamma-ray lines has been demonstrated[8] to be capable of ~ 1% assay accuracy in MSBR fuel samples, using appropriate standards (cf. Table V); however, in this case passive assay is time consuming and subject to some ambiguities.[9]

TABLE IV

INTERCOMPARISON OF DELAYED NEUTRON ASSAY OF MSBR FUEL WITH WET CHEMISTRY ASSAY[7]

ORNL Sample No.	^{232}Th Weight (grams)	Wt. % of ^{232}Th	
		LASL	ORNL
6A	13.80	Standard	34.9
7	18.19 ± .13	45.6 ± .3	45.0
8	16.33 ± .11	40.9 ± .3	40.6

LASL Standard No.	^{232}Th Weight (grams) Measured by Delayed Neutron Yield	Known ^{232}Th Weight
R1	Standard	16.85 ± .01
R2	18.91 ± .13	18.98 ± .01
R3	21.12 ± .15	21.44 ± .01
R4	24.02 ± .20	24.25 ± .01

TABLE V

PASSIVE ASSAY OF MSBR FUEL[8]

(Based on the 2.61 MeV Gamma-Ray[a])

ORNL Sample No.	^{232}Th Weight (grams)	Wt. % of ^{232}Th	
		LASL	ORNL
6A	13.80	Standard	34.9
7	18.19 ± .13	44.4 ± 0.3[b]	45.0
8	16.33 ± .11	40.4 ± 0.3[b]	40.6

[a]Additional results of similar accuracy have been obtained by using some of the lower energy gamma rays emitted in the thorium chain.

[b]Error based on statistics only.

A second series of MSBR fuel samples, containing representative concentrations of both ^{233}U and ^{232}Th, will be supplied by ORNL to LASL in early 1969. Although the MSBR samples assayed thus far have been one-component systems (containing only the single fissionable species, ^{232}Th), the use of "tailored" energy spectra in neutron interrogation (cf. later discussion of tailored spectra) as well as delayed neutron kinetic analysis should provide good discrimination between the ^{233}U and ^{232}Th present in actual MSBR fuels. The ultimate goal of the MSBR work at LASL is to develop delayed neutron and associated analysis techniques for continuous, on-line nondestructive assay of "hot" MSBR reactor fuels.

Neutron interrogation and delayed neutron response methods for rapid nondestructive assay of FFTF fuel pellets and LMFBR fuel

prototypes have been developed and are currently being tested at LASL.

B. Small test samples of various fissionable material mixtures, compounds, etc. are being assayed using a variety of techniques at LASL. Under an AEC program of detailed intercomparison between chemical analyses and nondestructive assay techniques, test samples obtained in the course of AEC safeguards inspection visits to various U. S. plants are being assayed by delayed neutron response methods and the results compared directly with conventional chemical analyses of the same samples at the AEC's New Brunswick Laboratory. Some typical preliminary results[10] obtained in this intercomparison program are summarized in Table VI.

C. Fissionable Material Scrap. An important practical problem in special nuclear material accountability is the determination of the amount of fissile material in common scrap containers. Delayed neutron yield and kinetics measurements offer a promising method for quantitative scrap assay. Basically, the method consists of irradiating the scrap container with a modulated neutron source and then measuring the delayed neutrons resulting from the induced (n, F) reactions in the fissile material. Preliminary measurements have indicated sensitivity for detecting small (gram) quantities of fissile material in a standard 55-gallon steel barrel.

LASL's experimental program in fissionable scrap assay has been guided by computer simulation studies of delayed neutron and gamma-ray

TABLE VI

ASSAY OF SAFEGUARDS INVENTORY SAMPLES

FROM NUCLEAR MATERIALS AND EQUIPMENT CORPORATION[10]

Sample Description (Numec No.)	Sample Weight (g)	Delayed Neutron Nondestructive Assay (LASL)			Chemical Assay (New Brunswick)	
		Total U Weight (g)	g U/g sample	% ^{235}U	g U/g sample	% ^{235}U
$UO_3 + ZrO_2$ Binary (#3619)	1.921	1.44	.750	98.6	.7678	97.671
$UO_2 + ZrO_2$ Powder (#3941)	1.827	1.40	.766	96.4	.7545	97.666
$UO_2 + ZrO_2$ Fired Scrap (#5353)	2.045	1.56	.763	97.7	.7695	97.653
$UO_2 + ZrO_2$ Scrap Powder (#1611)	2.011	1.52	.756	96.0	.7444	97.665
UO_2 Green Scrap Pellets (#9332)	4.170	3.60	.863	20.2	.8678	20.059
UO_2 Scrap Pellets (#1765)	4.072	3.60	.884	16.0	.8794	15.968
$UO_2 + U_3O_8$ (#5373)	1.988	1.67	.840	9.91	.8467	9.409
UO_2 Blended (#6440)	2.046	1.82	.890	4.25	.8713	4.158

response to active interrogation of small amounts of fissile material interspersed in large amounts of various scrap materials.[11] These parametric computations, summarized in Figs. 5 and 6, point up the very useful general result that barrels (55-gal. size, commonly used for scrap storage) containing almost any common scrap material other than hydrogenous compounds, are effectively transparent to fast neutron interrogation and delayed neutron emission (cf. Fig. 5). Thus assay for fissile material is essentially independent of composition of the matrix in which the fissile material is interspersed. The case of a hydrogenous matrix material is the most difficult for neutron assay applications, but even here calculations have shown that very effective fissile-material assay can be carried out by the use of a moderator-reflector together with appropriate ("add-a-gram") normalization of delayed neutron response.* The resulting dramatic improvement in delayed neutron response for bulk hydrogenous systems is clearly illustrated by comparison of Figs. 5 and 6.

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*The add-a-gram method consists essentially of a direct experimental determination of the delayed neutron response of a small amount (nominally one gram) of each fissile species of interest as measured directly in the neutron energy spectrum of the actual system under interrogation. This is accomplished by irradiating a gram of material in closest possible proximity (internally or peripherally) to the unknown sample, and then rapidly transferring the gram of material to a shielded 4π delayed neutron detector. This irradiation-counting sequence is repeated as often as required to obtain desired counting statistics for the add-a-gram calibration. Further details of the add-a-gram method are given in Ref. 12.

3. Detector and Instrumentation Development

For both passive and active methods of nondestructive assay it is important to have detection systems with the highest possible detection efficiency for both neutrons and gamma rays. In the case of passive assay, high efficiency (and often high resolution, as well) is necessary because of the characteristically low intensity of passive emanations. For active interrogation, high detection efficiency is required to maximize sensitivity and signal-to-noise characteristics while minimizing the strength of the interrogating neutron source and thus reducing the associated shielding requirements.

For many neutron counting applications, it is desirable to use a detector which has, in addition to high efficiency, a response independent of neutron energy. Two types of detectors satisfying these requirements have been developed and used extensively for safeguards research and development.[13] The first is essentially a modified Hansen-McKibben long counter consisting of five 1.5"-diameter ^{3}He (6 atmos) proportional counters placed in a 9-in. diameter polyethylene moderating cylinder. This detector has a neutron-energy response which is as flat as that of the original Hansen-McKibben long counter (which employs a single BF_3 tube) and an absolute efficiency which is about 50 times greater.

The second detector, which has a response almost independent of energy and an intrinsic detection efficiency of ~ 18%, is the "N-6 slab

detector." This detector consists of thirteen ^{3}He counters (6 atmospheres, 1" O. D. by 20" long) imbedded in two 2" x 20" x 24" slabs of polyethylene which are positioned in tandem. The counters in each slab may be operated as independent detection units or all thirteen counters may be connected in parallel, feeding into a single preamplifier. A special high stability FET preamplifier developed by the LASL Safeguards Group[14] permitted the operation of all counters in parallel with an overall pulse height resolution not significantly different from that of a single counter. Additional neutron moderating and absorbing material may be inserted in front of and between the slabs in order to vary the energy dependence of the efficiency of the detector.

A cross-sectional view of the N-6 slab detector in its "energy discrimination" mode is shown in Fig. 7. In this mode the ratio of the response of the front counter bank to that of the back counter bank provides a sensitive measure of the "average" energy of the incident neutrons. A plot of this ratio as a function of neutron energy, determined with various neutron sources, is shown in Fig. 8.

Exterior features of the neutron slab detector are shown in Fig. 9. A plot of its detection efficiency versus neutron energy when operated in the "flat" (energy independent) mode is also shown in this figure. These slab detectors have proved extremely useful in delayed neutron assay, as well as for other detection applications such as coincidence detection of

^{240}Pu spontaneous fission in a background of (α, n) neutrons. It is noteworthy that for the passive detection of ^{240}Pu, these N-6 slab detectors present a significant advancement over previous neutron detector technology, e. g. the BF_3 counter units provided to the IAEA by the United States.

A 4π detector with an absolute neutron detection efficiency of $\sim 65\%$ has been constructed for use in assay applications requiring extremely high sensitivity (e. g., for small samples and/or low neutron source intensities).

In addition to the slab neutron detectors, two large slab-type gamma-ray detectors have been designed and built. Among other applications, these detectors will be used for gross gamma-ray counting and for coincidence counting of prompt fission gamma rays, thus providing an independent means of detecting fissions occurring within a sample undergoing neutron interrogation. These detectors should be particularly valuable for coincidence counting ^{240}Pu spontaneous fission in scrap containing PuF_4. In this case, the chance coincidence counting rate of "single" neutrons from the copious $^{19}F(\alpha, n)$ source effectively masks the coincidence counting rate of the spontaneous fission neutrons.

Large volume, high resolution Ge(Li) detectors, as well as NaI detectors, are used routinely for passive gamma-ray assay.

An evaluation of commercially available small computers for

on-line data acquisition and reduction is presently underway at LASL. Such equipment, including input-output and interfacing units, will be used as a part of in-situ and in-plant assay systems as well as in the prototype mobile isotopic assay laboratory (cf. description in section 6) being developed by LASL.

4. Development of Neutron Sources, Moderators, Shielding, etc.

The sources available for neutron interrogation studies are small, inexpensive, high-yield neutron generators (D-D and D-T reactions, pumped-tube or sealed-tube type), positive ion accelerators, and radioactive sources (e.g., ^{252}Cf) as well as reactor neutron beams. A typical compact, mobile neutron generator, "Accelerator I", is shown in Fig. 10. For all fast neutron sources, considerably variability of neutron bombarding energy may be obtained by: (1) using either the D-D reaction (2.5 MeV neutrons) or the D-T reaction (14 MeV neutrons); (2) degrading or "spectrum tailoring" of source neutrons by surrounding the target with appropriate scattering and/or moderating materials. A series of computational and experimental studies of neutron moderator configurations used in conjunction with available high-energy neutron sources (D-D, D-T, and ^{252}Cf) has demonstrated the surprisingly high degree of flexibility in source spectrum which is attainable by spectrum tailoring methods. Some calculated tailored spectra for 14 MeV (D-T) and fission spectrum (^{252}Cf) sources are plotted in Fig. 11, and pertinent spectral

data are summarized in Table VII.[15] It may be noted that a 10-cm thick shell of tungsten can shift nearly 90% of the 14 MeV source neutrons to energies below the ^{238}U fission threshold. Note also that essentially the same spectral shift can be realized for a fission spectrum source (e.g., ^{252}Cf) using either tungsten alone or three concentric shells of tungsten, beryllium, and polyethylene in thicknesses of 5, 5, and 2.5 cm, respectively.

TABLE VII

MODERATOR CONFIGURATIONS[15]

Moderator Configuration (Thickness in cm)	Neutron Source	Total Leakage	Fraction of Neutrons with $E_n < 0.1$ MeV	Fraction of Neutrons with $E_n < {}^{238}U$ threshold
CH_2 - 10	14 MeV	89.9%	9.9%	16.8%
$Pb/C/CH_2$ - 12/10/2.5	14 MeV	141.5%	61.7%	82.5%
$W/Be/CH_2$ - 8/2.5/2.5	14 MeV	114.6%	61.8%	80.1%
$W/Be/CH_2$ - 5/5/2.5	Fiss.	77.5%	69.2%	88.7%
W - 10	14 MeV	139.0%	15.2%	86.6%
W - 10	Fiss.	86.4%	16.2%	88.4%

Monoenergetic neutrons of variable energies (subthreshold and superthreshold) are provided by a Van de Graaff accelerator as the interrogating neutron source. Figure 12 shows a 3-MeV high-current Van de Graaff recently installed in the Nuclear Safeguards Research Laboratory at Los Alamos.[16]

Neutron energy variability (whether by spectrum tailoring or directly with a Van de Graaff source) is important in many practical assay applications. For example, in low-enrichment power reactor fuel elements the "special nuclear material" (i. e., fissile species such as ^{235}U, ^{239}Pu, or ^{233}U) may be masked by large quantities of fertile material (e. g., ^{238}U or ^{232}Th); however, the relative response of the sparse fissile material can be greatly enhanced simply by shifting the energy of the primary source neutrons to the range below the threshold energy for fission of ^{238}U or ^{232}Th. Thus by the use of "subthreshold" fast neutron interrogation, good penetrability is preserved and, at the same time, relatively small quantities of fissile material can be detected in the presence of much larger quantities of fertile material; this capability is clearly of practical importance in the quantitative isotopic assay of power-reactor fuel elements, rods, plates, pins, pellets, scrap, etc.

As an alternative to accelerator and generator neutron sources, a ^{252}Cf spontaneous fission source of suitable intensity (e. g., 4 mg, giving 10^{10} neutrons/sec) offers a very practical, maintenance-free neutron source in extremely compact form, and having the convenient, highly penetrating, fission-spectrum neutron energy distribution. A ^{252}Cf neutron-interrogation assay system is presently under development at LASL.[17]

5. Intercomparison of New Nondestructive Assay Methods with Conventional Chemical Analysis

Before a given nondestructive assay method is adopted for a particular field application, it must be fully proved-in by detailed and extensive intercomparison with standard chemical (destructive) assay techniques. Such a program of intercomparison is underway at LASL in the areas of reactor fuel and fuel element analysis, small sample analysis, and scrap assay.

In the area of fuel element analysis, the delayed neutron method of assay on Rover fuel elements is being compared with chemical assay on portions of the same elements at the AEC's New Brunswick Laboratory. As already mentioned, MSBR fuel salts are being analyzed by nondestructive assay methods at LASL and compared with direct chemical assay at Oak Ridge. Also, delayed neutron techniques being developed for assay of FFTF fuel pellets and LMFBR prototype fuels will be compared with established chemical assay techniques used at Los Alamos. Nondestructive assay methods under development at LASL for low-enrichment water-moderated power reactor fuel elements (both cold clean and hot "spent" elements) will also be compared with chemical assay results from several laboratories.

In the area of small sample analysis, a number of test samples of uranium- and plutonium-bearing compounds and mixtures in various

physical and chemical forms are being analyzed nondestructively by both passive and delayed neutron methods at LASL for direct comparison with chemical assay on the same sample materials at the AEC's New Brunswick Laboratory (cf. results on Numec samples, Table VI).

In the area of fissionable material scrap analysis, nondestructive assay is being carried out on Rover scrap material (largely graphite plus 93% ^{235}U), and on typical scrap configurations and containers generated in the ^{235}U and Pu processing facilities at Los Alamos. These assay results will be compared with known scrap standards prepared at LASL and, whenever possible, with established destructive assay techniques long in use at LASL.

6. Adaptation of New Assay Methods to In-Plant and In-the-Field Systems for Practical Use by AEC and the Nuclear Industry

The techniques for direct on-line nondestructive assay of fissionable materials being developed at Los Alamos are directly applicable not only to safeguards but to nuclear materials management and accountability problems throughout the nuclear industry. Most of the nuclear industry is unaware of the importance and economic impact which the new nondestructive assay techniques can and will have on their operations, but an ever-increasing number of research-oriented people in industry and elsewhere are eager to explore the full potential of these new techniques as applied to a wide range of practical in-plant and in-the-field problems.

In discussions between LASL and OSMM officials, as well as with representatives of industry (the Atomic Industrial Forum, major fuel reprocessors and fabricators, etc.) it seems generally agreed that a most effective way of communicating and demonstrating these new techniques to industry is to field a demonstration unit, such as a roadable van, containing both in-place and portable assay instrumentation. Such a mobile assay unit, employing passive methods as well as active interrogation, would serve both as an assay laboratory per se, and as a staging area for conducting in-plant assay using portable instrumentation carried in the mobile unit.

A prototype mobile assay laboratory as envisaged by LASL is shown schematically in Fig. 13. The indicated neutron source for active interrogation can be a commercial pumped-tube type neutron generator, now available with all necessary beam pulsing and beam gating equipment, and capable of fast, convenient target replacement. As an alternative neutron source providing maximum compactness, replacement-module design and minimum maintenance, the newest commercially available sealed-tube-type neutron generators are currently being evaluated by LASL for nondestructive assay applications under both laboratory and field conditions.

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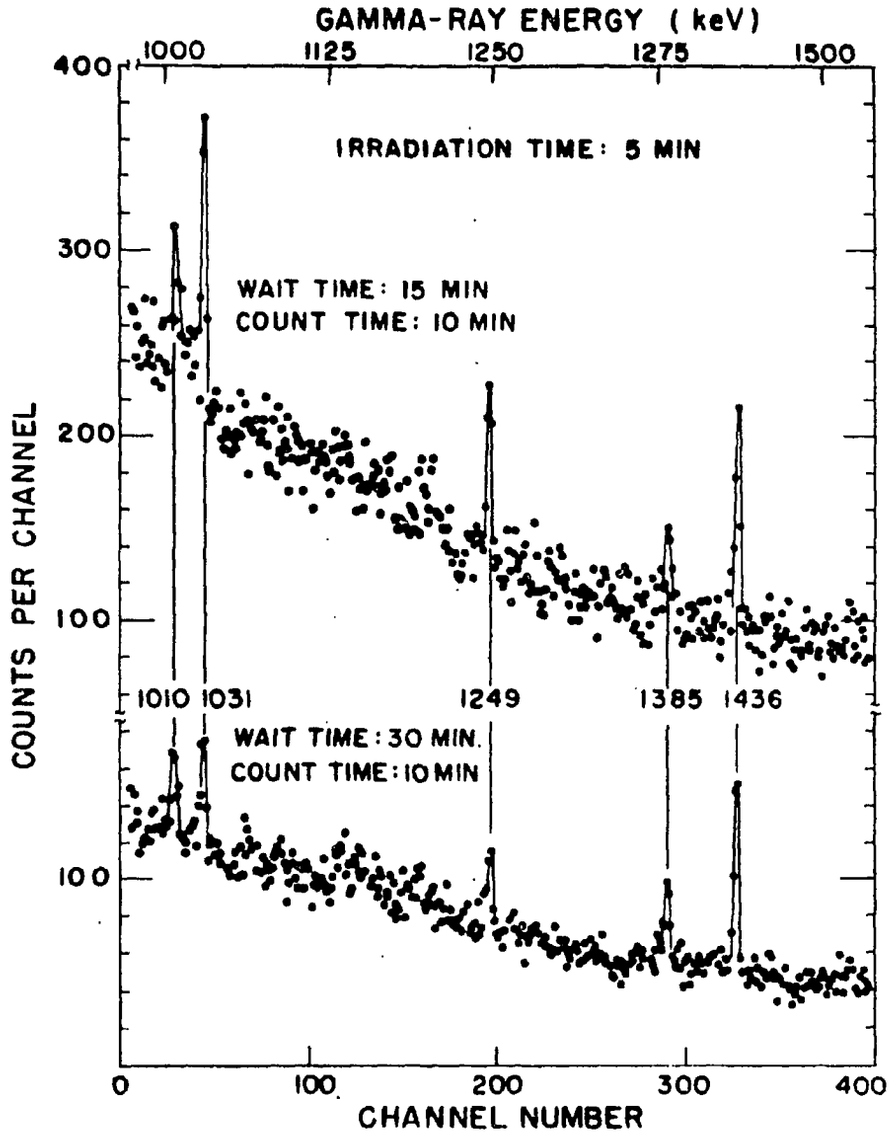


Fig. 1. $^{235}U(n, F)$ delayed gamma-ray spectra in the interval from 1.0 to 1.5 MeV at 15 and 30 minutes following a 5-minute irradiation of a 0.7-gm ^{235}U sample. The spectra were obtained with a 20-cc Ge(Li) detector and a one-inch Pb absorber interposed between the sample and detector. The 1.436-MeV line is tentatively identified with the decay of the 32-min. fission product, ^{138}Cs.

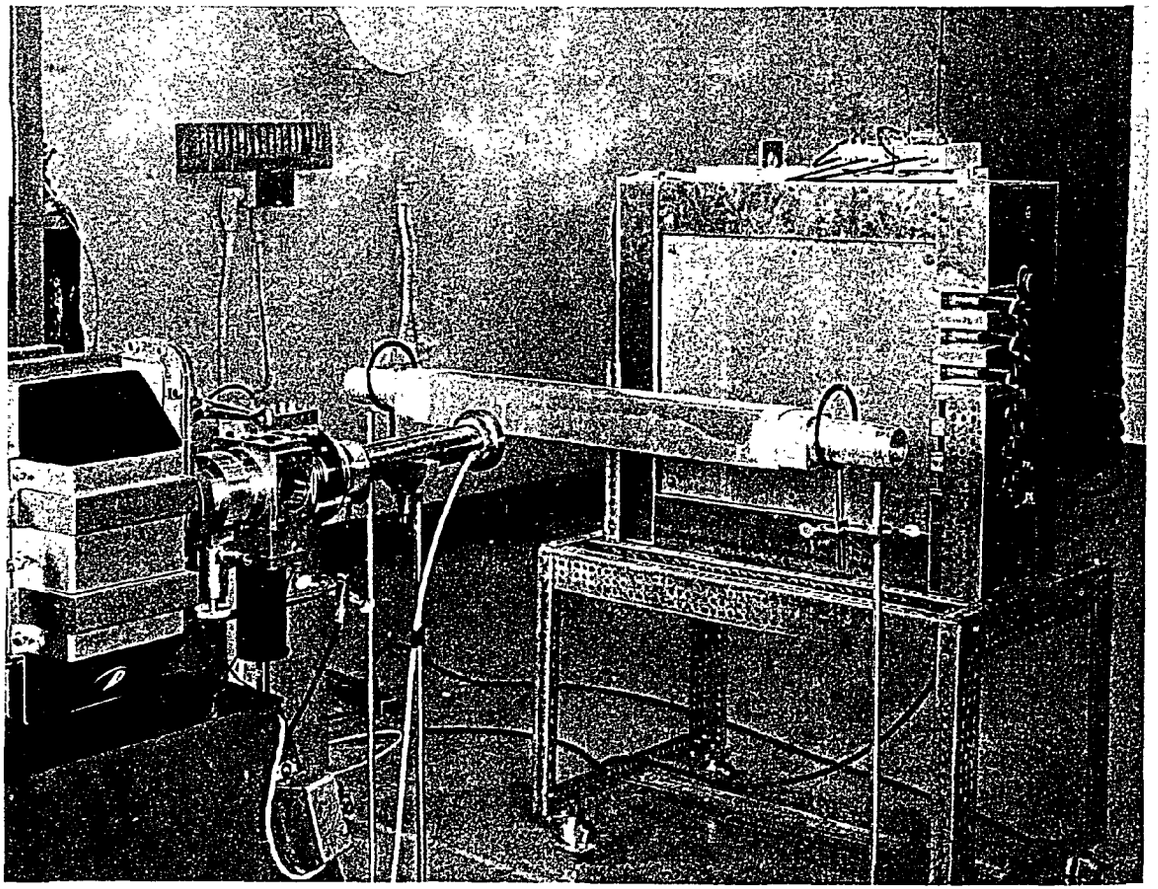


Fig. 2. An experimental arrangement for delayed neutron assay of bare MTR-type reactor fuel elements. The neutron interrogation source is seen at the left and a high-efficiency neutron detector, for measuring delayed neutron response, is seen behind the MTR element. Assay accuracies of 0.5%-1% are attainable using this method of interrogation.

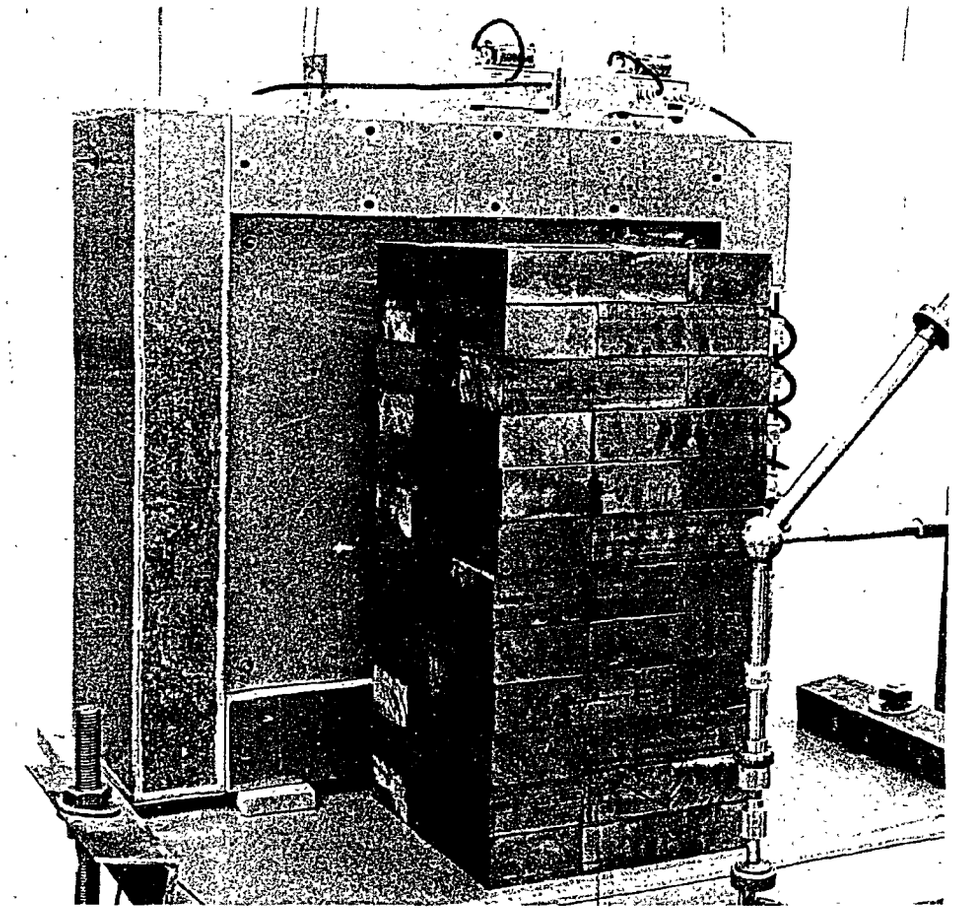


Fig. 3. Experimental arrangement for delayed neutron assay of MTR-type fuel element completely enclosed in a lead shield. The D-T neutron source is in the stainless steel sphere seen in the foreground, and the high-efficiency neutron detector is located behind the large lead shield.

Fig. 4. Solidified test sample of MSBR fuel salt (mixture of BeF_2, LiF, ThF_4, $^{233}UF_4$) showing "marble-like" heterogeneous solid sample for which representative chemical assay is extremely difficult. Delayed neutron assay techniques now provide a new direct method of assay nondestructively, with accuracies better than 0.5% (see text).

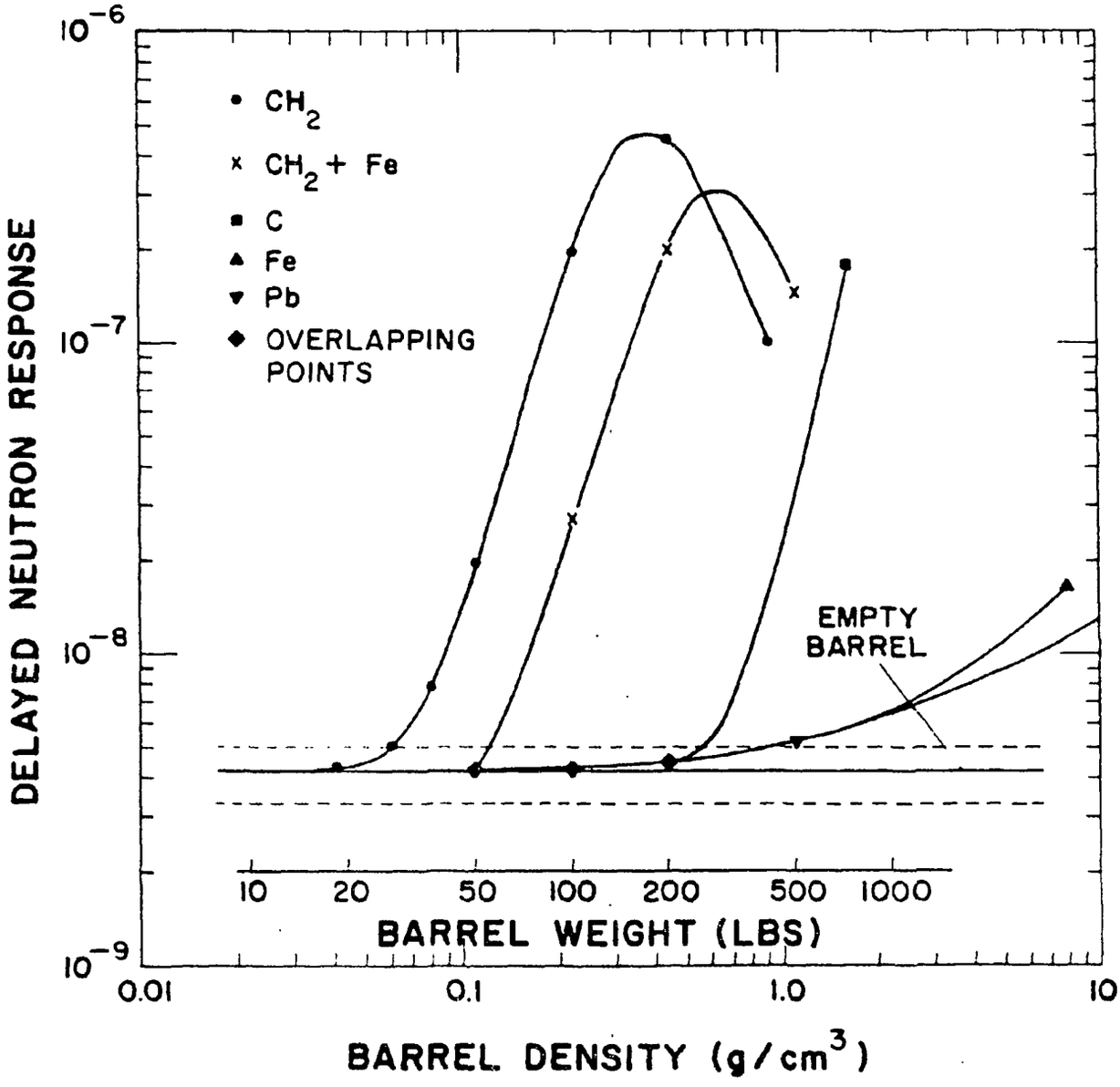


Fig. 5. Delayed neutron response of a simulated scrap barrel containing one gram of ^{235}U at 15 cm in representative moderator or matrix materials--14-MeV source. The dashed lines indicate empty barrel (no moderator) response ± 20%. (The ± 20% error span represents reasonable accuracy limits for practical scrap assay.)

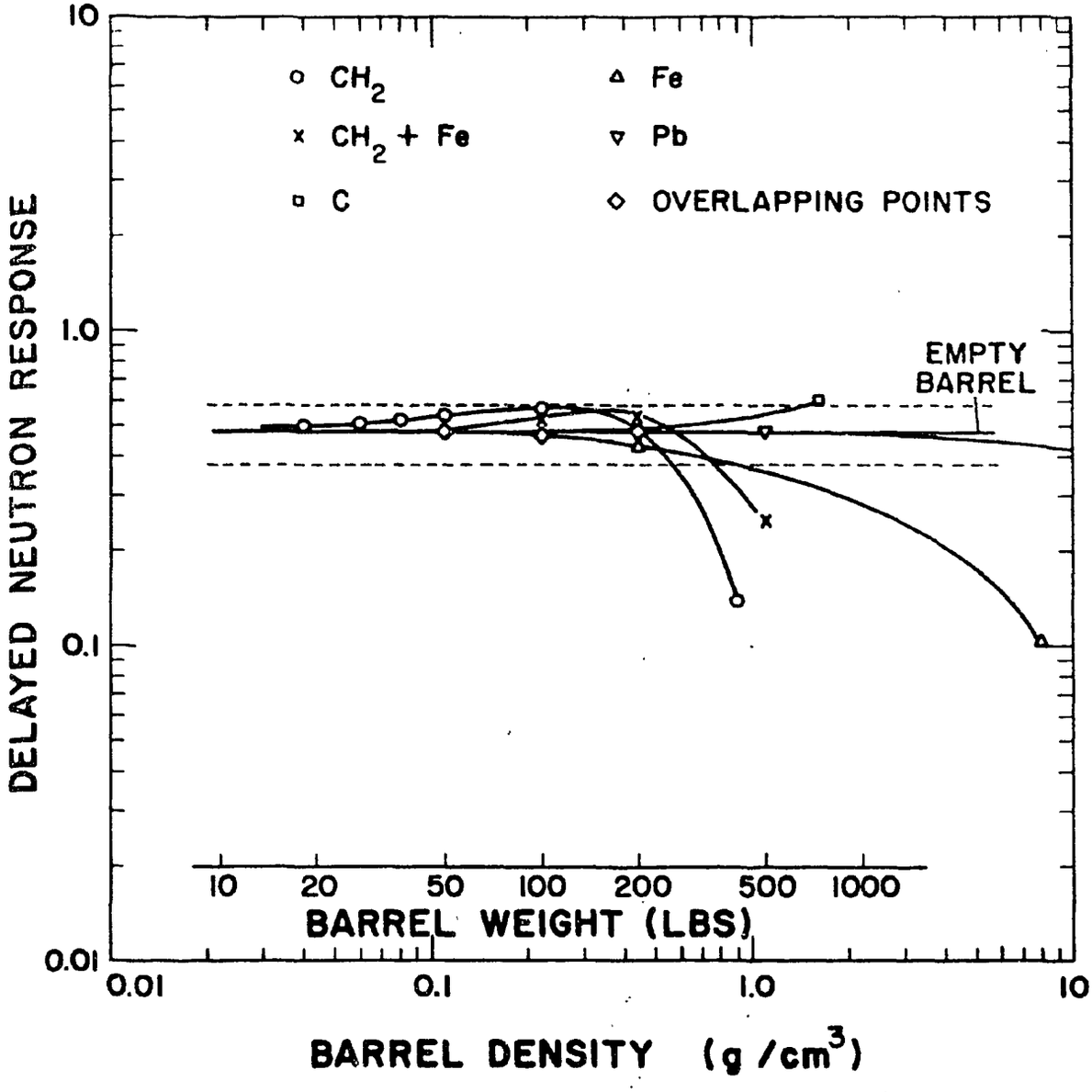


Fig. 6. Delayed neutron response of a simulated scrap barrel surrounded by a 2.5-cm polyethylene reflector. The response has been normalized to the "add-a-gram" (see text) response. The dashed lines indicate empty barrel (no matrix material) response ± 20%.

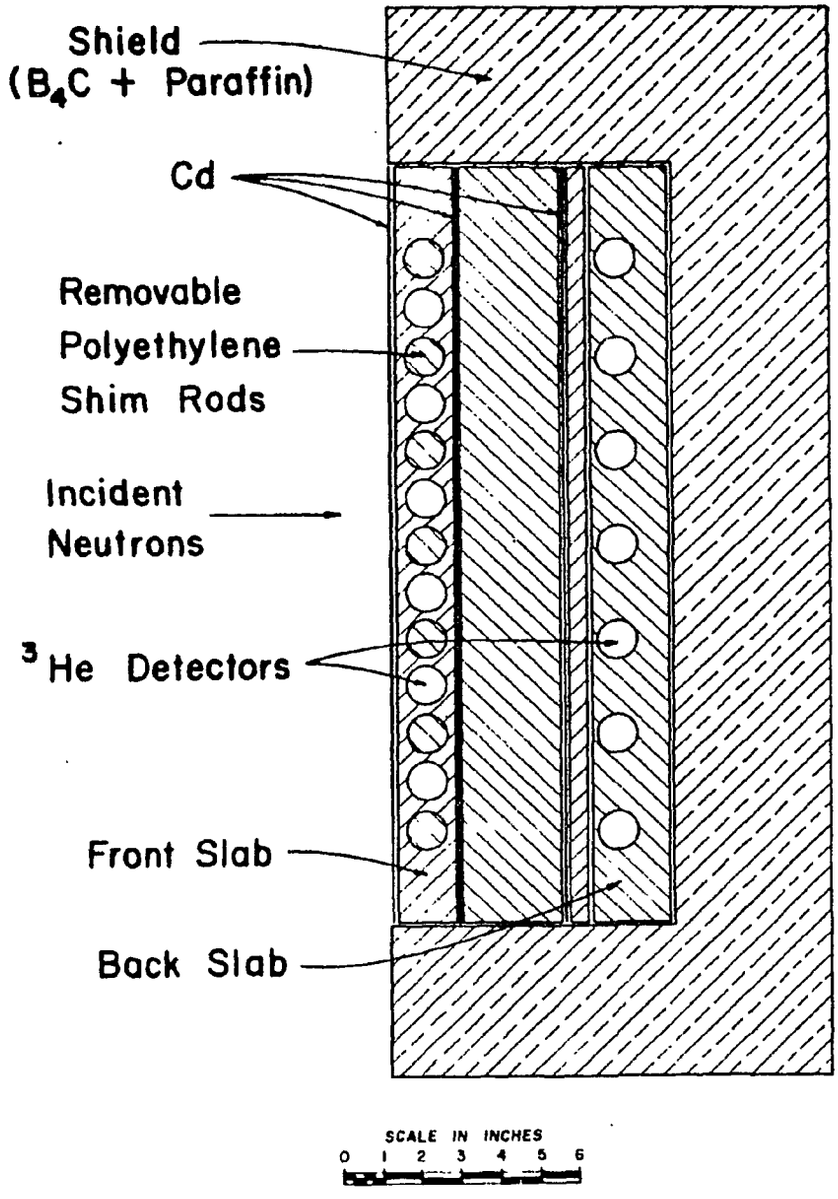


Fig. 7. Cross-sectional view of the N-6 slab detector. This configuration is used to obtain coarse neutron-energy spectrum information. The detector may be converted to its flat response mode of operation by removing the inner Cd pieces and rearranging the polyethylene moderator.

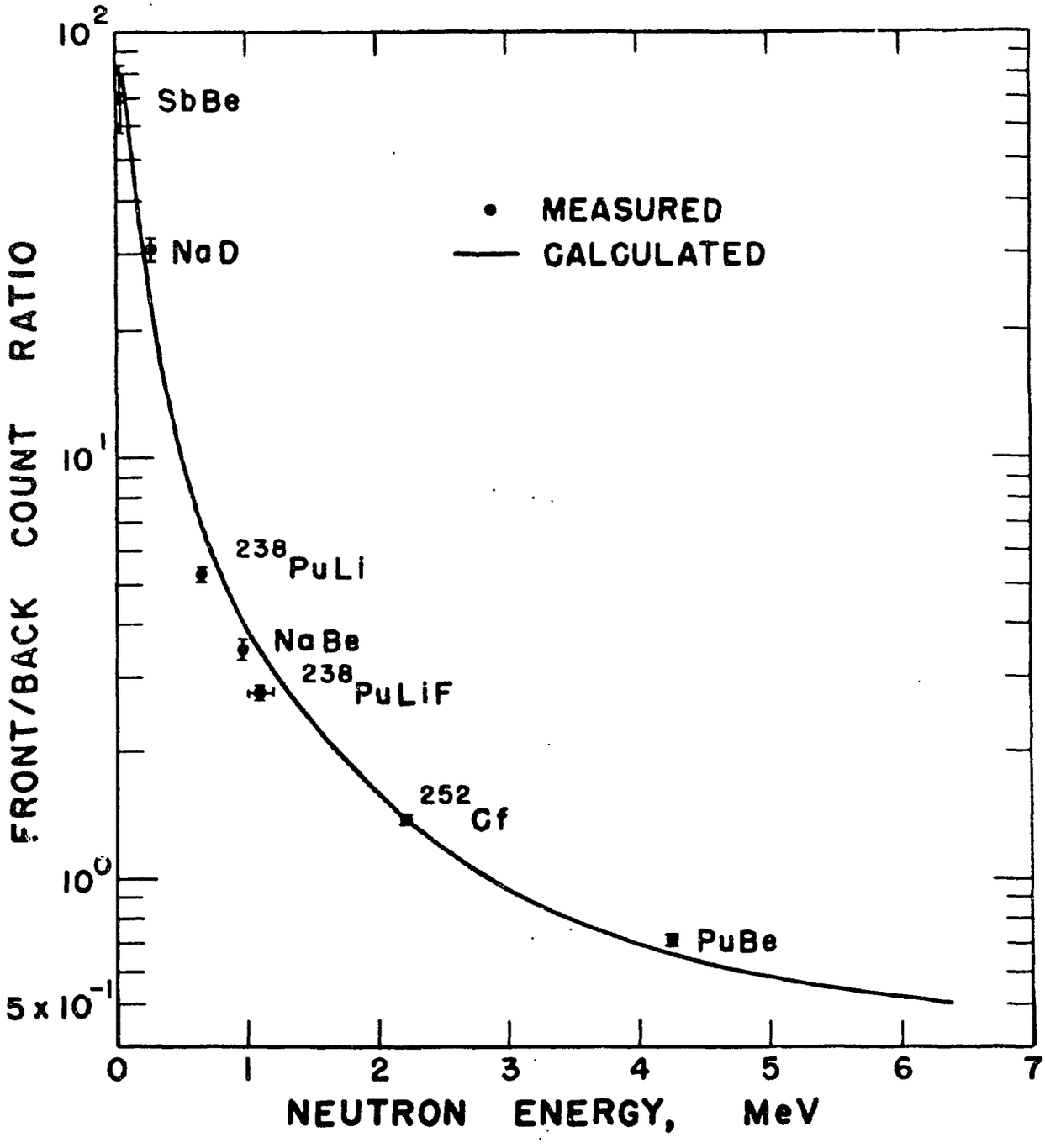
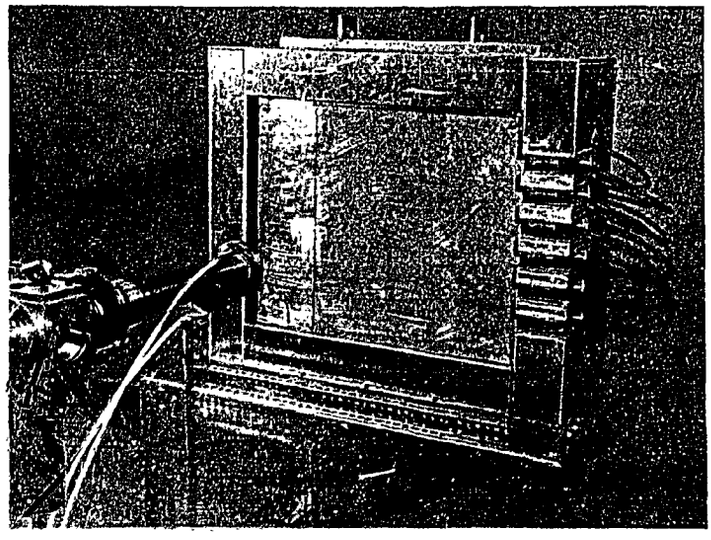


Fig. 8. Response ratios between front and back counter banks of the N-6 slab detector versus incident neutron energy. The points (with error bars) represent measured results, and the smooth curve shows the front/back ratio calculated using a one-dimensional transport code.



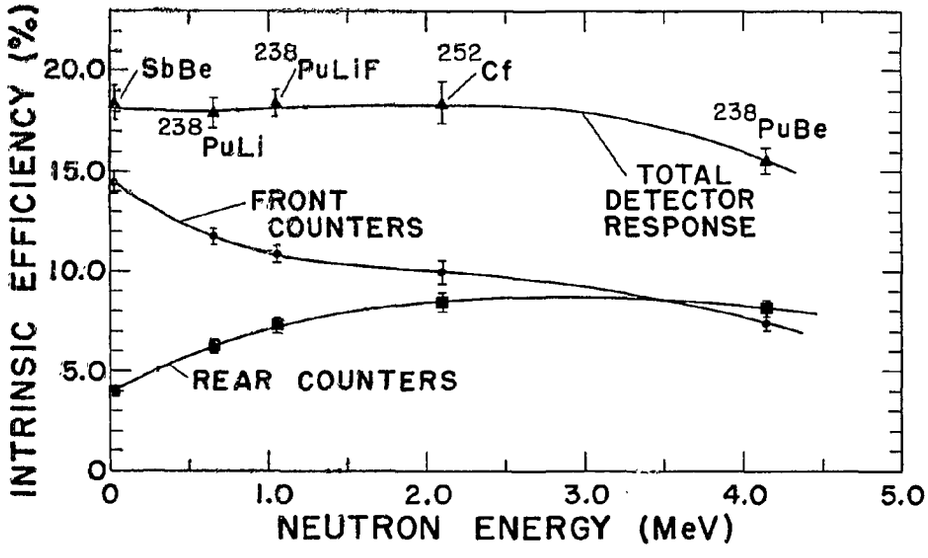


Fig. 9. The photograph shows an N-6 high-efficiency slab detector having 18% intrinsic efficiency over a sensitive area of 20" x 24". The Accelerator I neutron source (beam target shown in left foreground) in combination with the slab detector is used extensively for delayed neutron assay techniques being developed at LASL. The lower portion of the figure shows the intrinsic efficiency of the detector versus neutron energy; the indicated flat response as a function of energy for the sum of the front and back counter banks (i. e., the total detector response) is necessary for most applications involving assay of large complex arrays of nuclear material such as fuel elements, nuclear devices, scrap, etc.

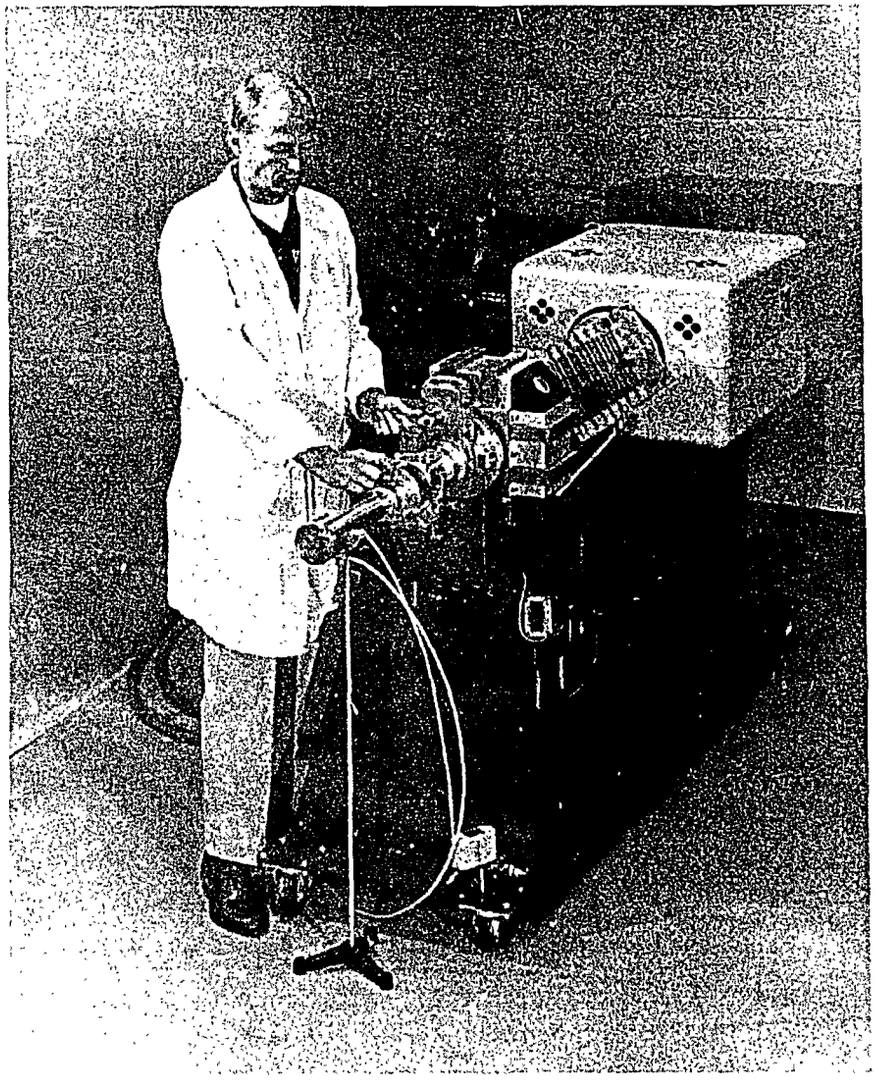


Fig. 10. Accelerator I - A Compact, High Intensity Source for Neutron Interrogation and Nondestructive Assay. (Accelerator potential: 150 kV; maximum beam current: 3.5 ma; beam pulsing by pre- and post-deflection; beam modulation by gating the RF ion source.) Maximum source intensity is $\sim 3 \times 10^{11}$ D-T neutrons/sec; the source can be gated on and off as required for optimum neutron interrogation applications. Two new quick-change target holders have been adapted to Accelerator I for fast, convenient target replacement in laboratory and field installations.

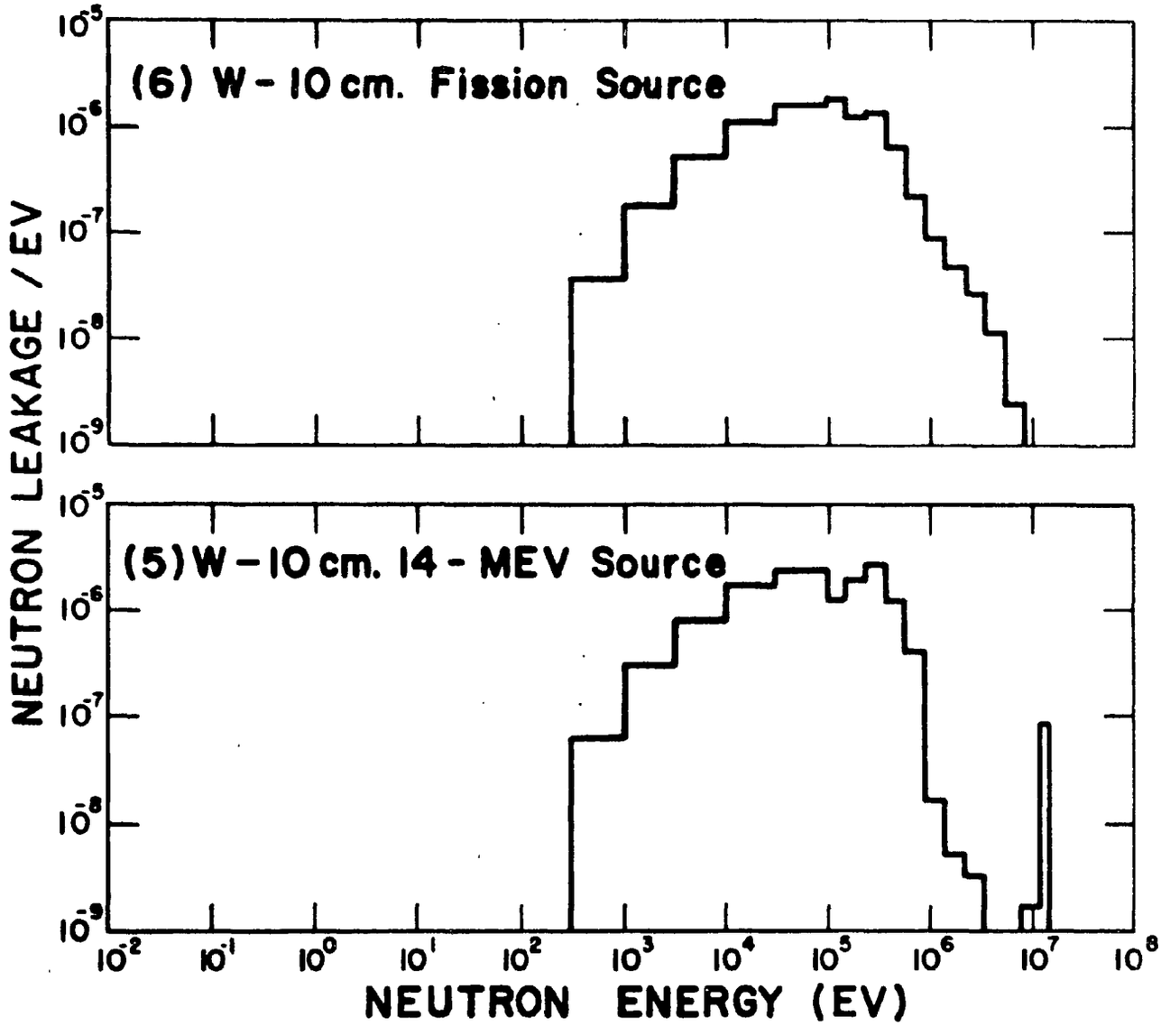


Fig. 11. Neutron leakage spectra from a tungsten "fast moderator" assembly surrounding both 14 MeV and fission-spectrum neutron sources. (Ordinate normalized to one source neutron.)

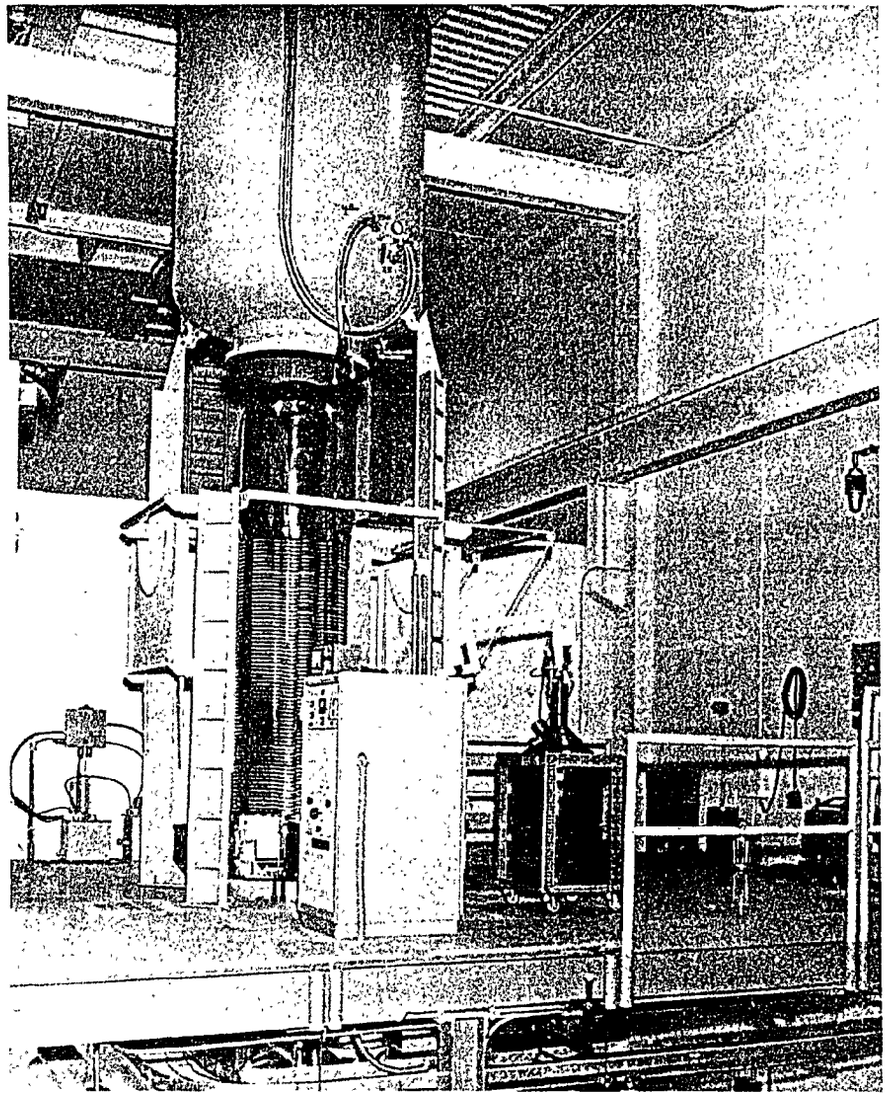
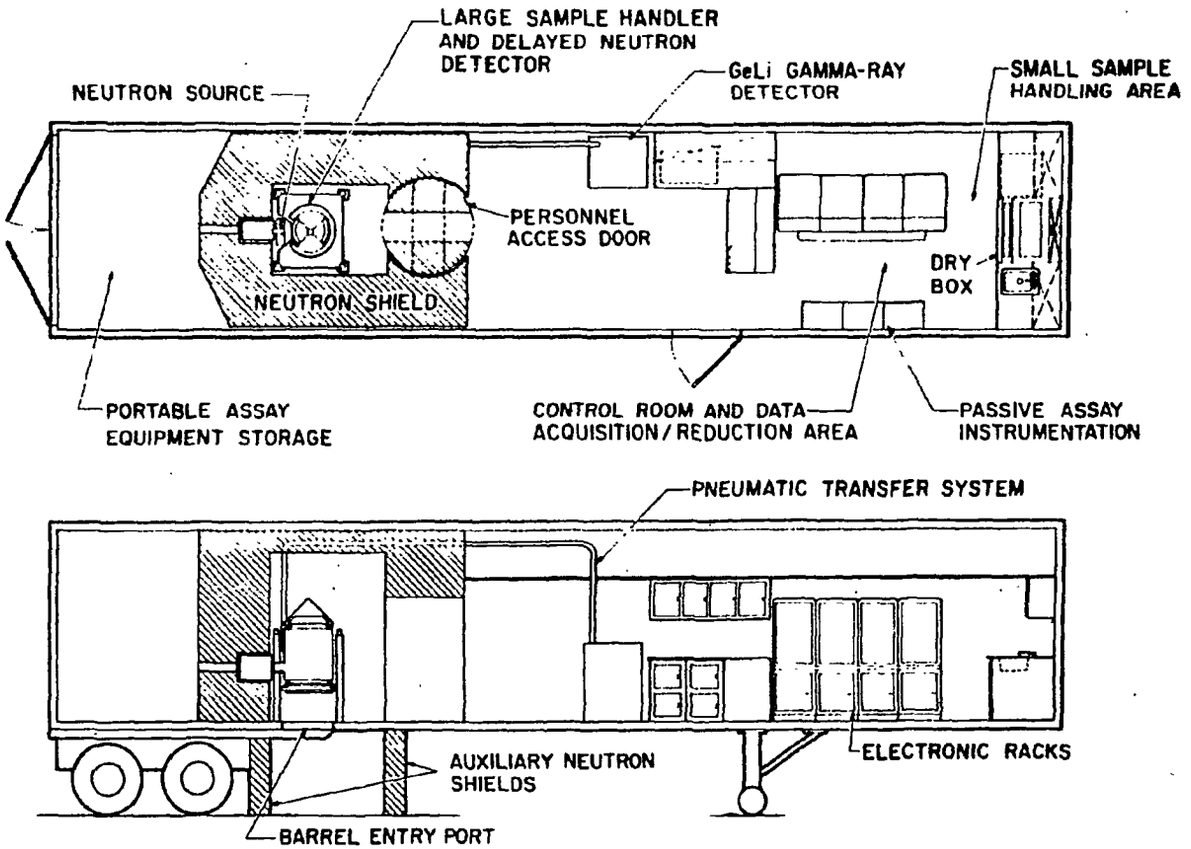


Fig. 12. General view of the 3-MeV high-intensity Van de Graaff accelerator installed in the Nuclear Safeguards Research Laboratory at Los Alamos. (High Voltage Engineering Corporation KN-3000 Vertical Machine, 750 μa analyzed beam of protons.) Monoenergetic neutrons of variable energies (both above and below the "threshold" energies required to fission ^{238}U, ^{232}Th, and ^{240}Pu) are used to interrogate fuel elements and other fissionable material systems and devices. Using appropriate standards, nondestructive isotopic assay by neutron interrogation techniques can be carried out in many practical cases to better than 1% accuracy.



MOBILE SAFEGUARDS ASSAY LABORATORY

Fig. 13. Schematic layout of mobile isotopic assay laboratory currently under development at the Los Alamos Scientific Laboratory. The mobile laboratory is incorporated in a 10' x 50' cargo container (roadable van) which can be transported by land, sea, or air. Active interrogation (of systems as large as a 55-gallon barrel) will be performed in the shielded area at the rear (left) of the van. The front of the van serves as control room, data analysis station, and a general laboratory for passive assay of fissionable materials.