THE EXPANDING ROLE OF THE SMALL VAN DE GRAAFF IN NUCLEAR NONDESTRUCTIVE ANALYSIS*

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Summary

In support of Nuclear Safeguards, a 3-MeV Van de Graaff accelerator was installed at LASL to aid research in techniques for nondestructive assay of fissionable materials. Monoenergetic neutrons make possible fissile assay techniques less influenced by self-absorption than thermal interrogation and more discriminating of fissile over fertile material than use of partially moderated high-energy neutrons. Delayed fission-neutron counting, a valuable assay technique, is facilitated by pulsed neutron on-to-off ratios in excess of 10^9, achieved with special beam-handling techniques. Where high neutron backgrounds, e.g. from samples containing ^{240}Pu, preclude delayed-neutron counting, fission prompt-neutron counting with an energy-biasable detector is applied. The increasing facility workload, which includes detector development and calibration, research in trace analysis by proton-induced x-ray fluorescence and research in light-element isotopic assay by means of charged-particle induced reactions, indicates that technological application may more than supplant dwindling nuclear research as a market for accelerators of this class.

Introduction

In the nuclear physics research laboratory, the small Van de Graaff has almost become an orphan. Federal support of these machines in university, government, and nonprofit laboratories has been drastically cut. Even at Los Alamos in the period prior to 1967, three 3-MeV machines, no longer relevant to the research needs of the laboratory, had been decommissioned. Then, in January of 1967, there was initiated at Los Alamos a program of research in nondestructive detection, identification, and analysis of fissionable materials in support of national and international Nuclear Safeguards.[1] It was evident from the start that many problems in nondestructive fissionable material assay could best be solved by interrogation with fast neutrons.

Neutron Sources for Safeguards Research

The selection of a neutron source for a nuclear assay system requires more than casual consideration. In the Los Alamos program we have used radioactive neutron sources,[2] small Cockcroft-Walton type (d, t) neutron generators,[3] and a reactor.[4] Each of these source types can play a significant role in nuclear materials assay. However, early in the program it was realized that in order to study the problems associated with the application of neutron interrogation to nuclear materials assay, a neutron source was needed which was variable in energy spectrum and intensity and which could be turned on and off at will. Accordingly, a 3-MV Van de Graaff accelerator was purchased in 1968 and installed in an unused reactor test facility building.

The accelerator installation is shown in Fig. 1. The machine is a standard HVEC vertical model KN-3000, which has since been upgraded to a maximum rating of 3.75 MV. Features which were nonstandard at the time of purchase include a 42 in. high terminal for future accommodation of complex ion-source systems and a voltage control system designed to operate either (1) from the output of the generating voltmeter, (2) from the current differential on control slits following momentum analysis, or (3) mode (2) switching automatically to mode (1) in the event of loss of control-slit current. The momentum analysis system will handle ions with mass-energy products up to twelve.

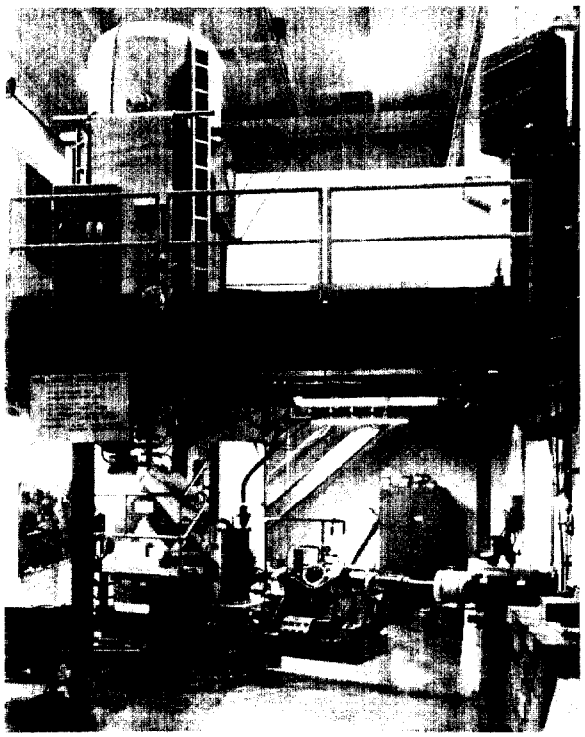

Fig. 1. The LASL 3.75-MeV Van de Graaff accelerator and beam analysis equipment.

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An Example: Small Sample Assay by Delayed-Neutron Counting

The Van de Graaff has provided us with our most effective method for assay of ^{235}U in small samples. Samples are interrogated with 300-600 keV neutrons, produced by bombarding 2 mg/cm^2-thick lithium targets with 2.35-MeV protons. The modulated source technique developed by Masters et al.[5] is used to measure the delayed-neutron yield from fission in the samples. Delayed neutrons, which account for 0.6% to 5% of the neutrons emitted in fission, are practically unique to the fission process and are very easy to count. Briefly the technique as now applied consists of repeatedly bombarding the samples in cycles consisting of a 35 msec neutron pulse followed by a 15 msec "wait" period, a 35 msec counting period, and another 15 msec "wait" period. It has been shown[5] that if the neutron bombarding cycle is operated for a period long with respect to the longest half-life of the delayed-neutron groups (which have half-lives of from 0.1-56 sec), then the yield from delayed neutrons is independent of the delayed-neutron group abundances and half-lives, and is proportional only to the quantity of fissile material present, the interrogating neutron flux, the fission cross section integrated over the neutron spectrum, and the absolute delayed-neutron yield per fission, the latter quantity being a constant for a given isotope in the interrogating neutron energy region of interest.

Fission chambers are used to monitor the interrogating flux. These are placed close to the sample so that one may at least partially compensate for flux-depression effects. The fission chambers are preferrably loaded with the fissile material for which one is assaying in order that neutron spectral shifts--such as those due to lithium target deterioration--will not alter the calibration of the system.

Counting statistics, the principal determinant of precision, are of course determined by the product of interrogating neutron strength, fissile content, and the length of time which one is willing to spend on each sample. The limit of sensitivity, however, is set by the ratio of neutron flux at the sample during the interrogating pulse to the background neutron flux in the delayed-neutron detector during the "off" period of the irradiation cycle. With the sample located 10 cm from the neutron source, calculations show that for the background to be equal to the delayed-neutron response from 1 g of ^{235}U, an on-to-off neutron ratio of $\sim 10^7$ is required. The "off" neutron count includes both a constant "ambient" time-dependent component and an accelerator-induced beam-dependent component, each of which must be treated separately in the analysis. In our setup, we find ambient backgrounds equivalent to the delayed-neutron response from $\sim$ 15 mg of ^{235}U for our best available source strength of $\sim 10^{11}$ n/sec. Beam-induced backgrounds are equivalent to from 6-8 mg of ^{235}U, indicating that we are achieving on-to-off ratios in excess of 10^9.

Beam Handling

Achievement of such an on-to-off ratio with a Van de Graaff beam requires some care in the planning of the facility and in the handling of the beam. Figure 2 shows the layout of the accelerator facility. Three feet of concrete shield the accelerator from the experimental area which is shown in Fig. 3. Because the time constant of the desired beam pulses is long with respect to the 2-3 msec R-C constant of the Van de Graaff column and terminal, it is not readily feasible to pulse the accelerator current, which is of the order of 100 μa before momentum analysis. For this reason, beam

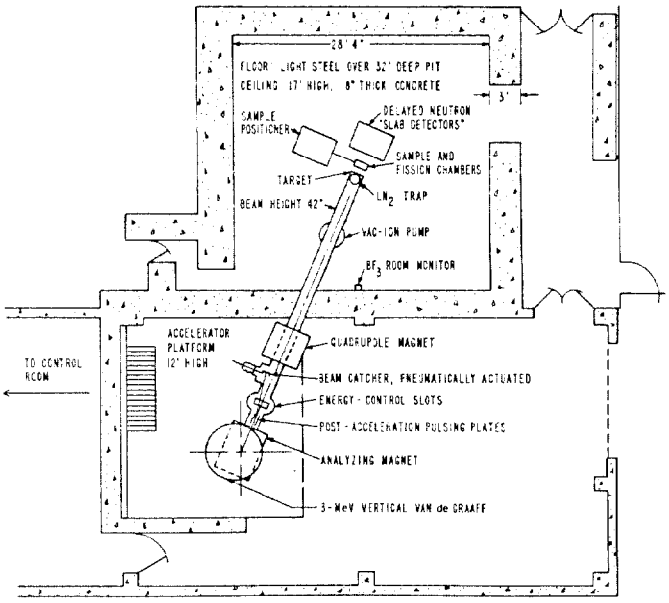

Fig. 2. Floor plan of the accelerator facility.

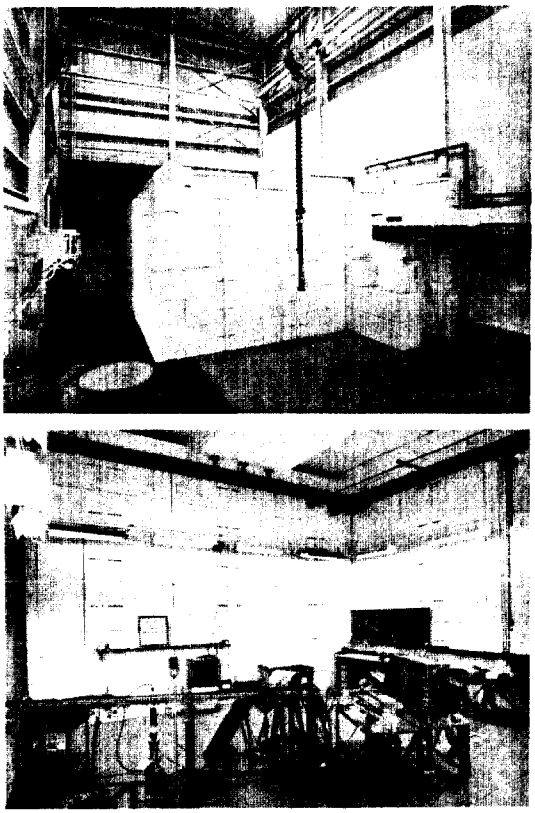

Fig. 3. Exterior (upper photo)and interior (lower photo) views of the shielded experimental area.

pulsing is accomplished after acceleration and momentum analysis, by means of a rotating mechanical shutter and synchronized electrostatic deflector on the beam line. The mechanical shutter is shown in Fig. 4. Figure 5 shows the electrostatic beam deflector, the water-cooled catcher which precedes the shutter, and local shielding used to suppress neutrons from the mass-2 port of the beam-analysis system, where most of the deuterons from the natural hydrogen beam are collected.

Effect of Low-Energy Neutrons

We initially tried small-sample assay by measurement of delayed-neutron yields using the geometry shown in Fig. 6. The delayed-neutron detector is the "slab detector" of East and Walton[6] consisting of 13 ^{3}He proportional counters imbedded in polyethylene. Tests were run using various quantities of uranium oxide, enriched to 93% ^{235}U, mixed with varying quantities of graphite and powdered aluminum in 1-dram glass vials which were selected as an interim standard container. Appreciable matrix-dependence effects were found; for example, a vial containing 1 g of uranium as oxide was found to have a delayed-neutron response 11% less than the same quantity of enriched uranium mixed with three times its volume of graphite. This problem could be partially circumvented by calibrating with standards which closely match the unknown, an impractical solution if one is dealing with a great variety of samples of unknown composition. Elimination of the cadmium shield about the sample and fission chambers made the system completely unreliable. It was finally found that insertion of a $\frac{1}{4}$"Boral plate between the slab detector and the sample noticeably reduced matrix dependence. This is shown in Fig. 7. The upper curve of the figure shows the dependence of the delayed-neutron response from 1 g of 93% enriched uranium mixed with various quantities of powdered graphite or aluminum, as a function of the volume fraction of U_2O_3. The middle curve shows the effect after adding the Boral sheet.

It was concluded that the matrix-dependence effects were due to the presence of epicadmium, resonance-region neutrons, principally reflected from the

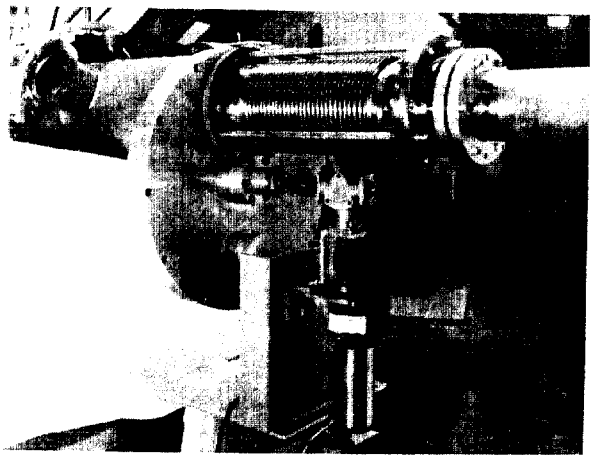

Fig. 4. Mechanical beam shutter shown installed on the horizontal beam line.

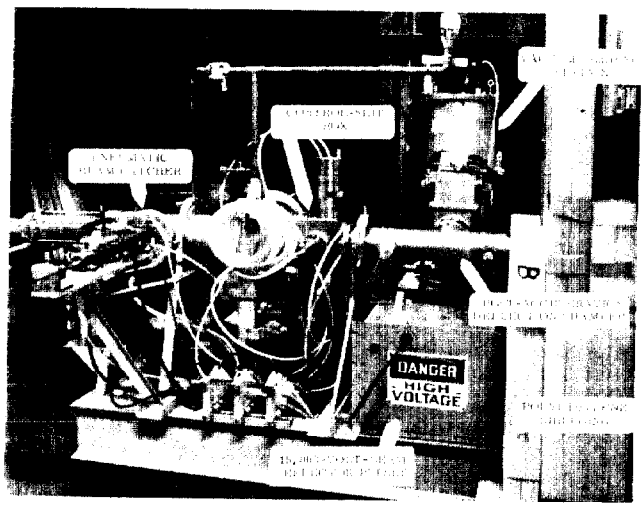

Fig. 5. Post-acceleration beam-deflection system.

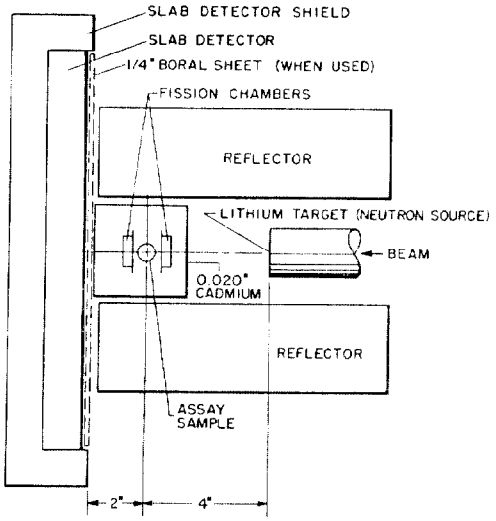

Fig. 6. Early setup for assay of small samples by subthreshold-neutron interrogation.

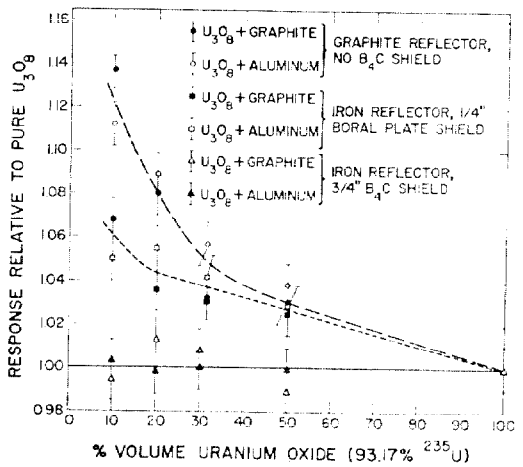

Fig. 7. Delayed-neutron response of 1 g of enriched uranium (93.17% ^{235}U) in various quantities of graphite or aluminum, using varying quantities of B_4C shielding.

detector, which caused the samples to exhibit large self-absorption effects. To eliminate this problem, a small-sample assay chamber was built with $\frac{3}{4}$ in.-thick hot-pressed B_4C walls. Using the B_4C shield, which is calculated to have an effective neutron cutoff of about 100 eV, the bottom curve is obtained. The assay chamber is shown in use in Fig. 8. The lower curve of Fig. 7 shows the improvement which was achieved.

It was thus demonstrated that precision assay requires a hard neutron spectrum, with a minimum low-energy "tail". Fissile-material assay systems based upon thermal-neutron interrogation, while more sensitive because of the higher fission cross section, will work only if standards are available which match the unknowns in every detail of geometry and chemical and isotopic composition. Naturally, clean separation of the responses of fissile and fertile materials requires that no neutrons in the interrogating spectrum for fissile isotopes be above the threshold for fission in the fertile isotopes.

Techniques for Other Nuclear Materials

Assay by neutron interrogation has been applied to fissionable materials in containers ranging from small vials to 55-gal drums. It has been shown to be feasible to determine the ^{235}U content of an entire BWR fuel bundle with an uncertainty of 2% or less.[7] The setup for this is shown in Fig. 9. In this case, both delayed and prompt-neutron measurements were made. The counting of prompt-fission neutrons was done with a bank of ^4He-recoil proportional counters, the output of which was biased to prevent counting of neutrons of energy less than 700 keV, thereby eliminating signals from the 200-500-keV interrogating neutrons. The ^4He recoil counters are sufficiently insensitive to gamma radiation that pulse-shape discrimination is not necessary in this application. Use of prompt, rather than delayed fission neutrons as an assay signature, is necessary when assaying such materials as recycle plutonium reactor fuel, which has a high background of (α, n) and spontaneous-fission neutrons. Measurement of prompt-to-delayed fission neutron ratios may eventually permit determination of Pu to ^{235}U ratios in reactor fuels. For the assay of fertile materials, such as ^{238}U or ^{232}Th, the interrogating neutron energy is raised above the fission threshold for these materials. For example, 2.0-MeV neutrons produced by 3.75-MeV protons on lithium are adequate for ^{238}U analysis.

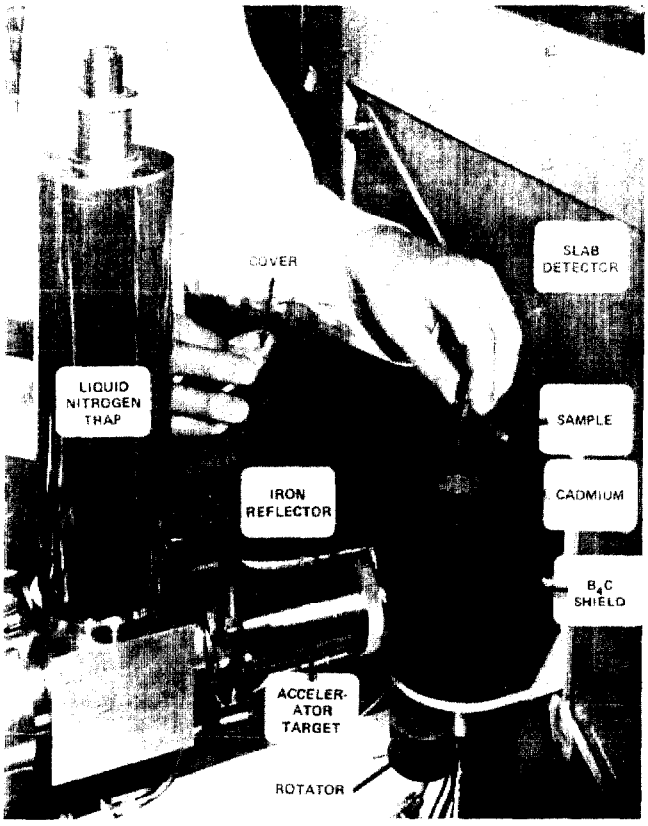

Fig. 8. Assay station with pressed-B_4C assay chamber.

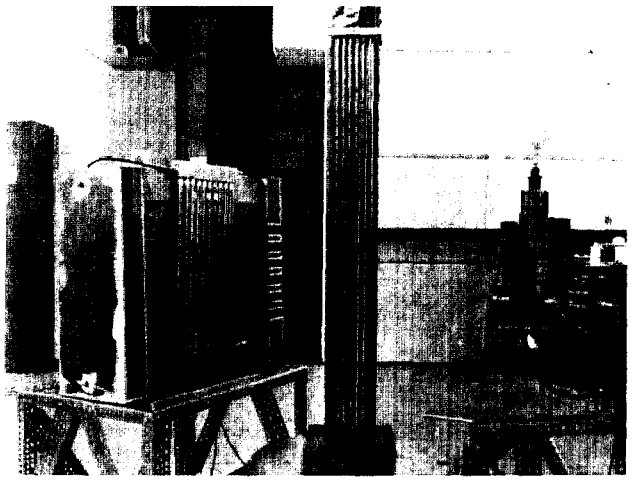

Fig. 9. Experimental arrangement for assay of power reactor fuel elements. The accelerator target is on the right. The ^3He delayed neutron slab detector and bare ^4He prompt-neutron detectors are on the left.

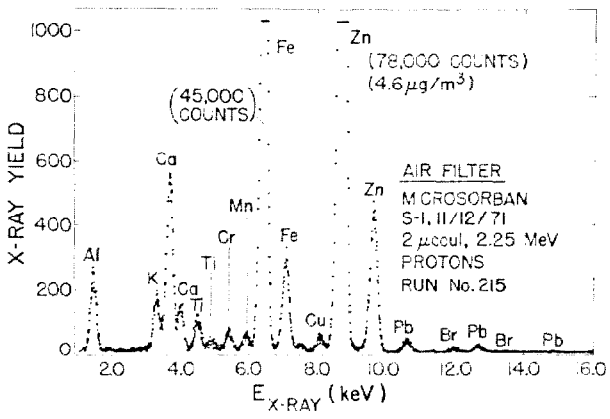

Fig. 10. Proton-induced x-ray fluorescence spectrum from a typical air-sampler air filter.

Other Analytical Applications of the Van de Graaff

Proton-Induced X-Ray Fluorescence

Another field in which a small Van de Graaff facility can be used is that of trace-element analysis by means of charged-particle-induced x-ray fluorescence. This technique is being used for extensive air pollution studies in California.[8] At LASL, we are investigating its application to local problems. It has been shown that this method of analysis is capable of detecting of the order of 10^{-11} g[9] of elements from sulfur to uranium, analyzing simultaneously for all elements with $Z \geq 15$. A typical example is shown in Fig. 10. This is of an air filter exposed for one week in an outdoor air sampler in Los Alamos--a reasonably clean environment. The accumulation of the spectrum took less than two minutes of running time using 2.25-MeV protons. The relatively high levels of zinc and iron were thought to have arisen from a welding operation in the neighborhood.

Charged-Particle Nuclear Reactions

We are currently investigating the feasibility of using radiative proton capture as a tool for isotopic analysis of light elements. There is at Los Alamos a capability for the production of kilogram quantities of such isotopes as ^{13}C, ^{15}N, ^{17}O, and ^{18}O. A method of analysis for these stable isotopes more convenient than conventional mass spectrometry would be of considerable benefit in making the isotopes more useful for biological tracer studies. Preliminary work on measuring ^{12}C: ^{13}C ratios has been reported elsewhere.[10]

Other Work

In addition to being an instrument applied directly to problems of chemical and isotopic analysis, the 3.75-MeV Van de Graaff has veen a valuable diagnostic tool for the development of instrumentation used in Nuclear Safeguards. The availability of monoenergetic neutrons to study the behavior of detectors as a function of neutron energy has been of inestimable value in optimization studies leading to the design of small, isotopic powered nuclear assay systems.[11] In addition to supporting our nuclear analysis program with such detector tests, we have been called upon by other groups in the laboratory to provide monoenergetic charged-particle and neutron beams to calibrate detectors ranging from TLD dosimeters for health physics application to charged-particle spectrometers for space exploration.

Conclusion

It appears that we have barely scratched the surface of application of machines of this type to man's industrial problems of materials analysis. A nuclear power industry which must grow to support the nation's energy needs will in turn create a need for nuclear analysis both for quality assurance and for materials control which can only be met by nondestructive assay. Growing concern over the harmful effects of small quantities of certain elements in our environment will generate a need for more and better facilities for trace-element analysis. Thus it is that the low-MeV particle accelerator, now almost a has-been in the nuclear physics research laboratory, may soon have a new and bigger role in the analytical laboratories of industry and government.

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