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CONF-731112--1

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SUBMITTED TO: 1973 IEEE Nuclear Science Symposium

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Form No. 836
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NONDESTRUCTIVE ASSAY OF FISSILE MATERIAL SAMPLES IN SUPPORT OF NUCLEAR SAFEGUARDS*

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Summary

Samples of fissile material can be assayed by bombarding with 300- to 600-keV neutrons and counting delayed neutrons from fission. Interrogating neutron energy selection is based upon considerations of sample penetrability and insensitivity of response to nonfissile isotopes. Significant cost savings in nuclear safeguards and quality control are possible.

Introduction

Concern over possible theft or unlawful diversion of fissile material from growing stockpiles in the nuclear power industry--with resultant unthinkable consequences--has caused a tightening of Federal requirements for the accountability of AEC contractors and licensees for this material. It is now proposed[1] that organizations having such material in their possession be able to ascertain at regular intervals (or continuously) their fissile inventories with sufficient accuracy that the measured limit of error of material unaccounted for (LEMUF) is no greater than ± 1/2% of their inventories. The achievement of such control requires an increased effort of measurement of all items in the fuel cycle: input feed material, in-process batches on line and in storage, finished nuclear fuel products, scrap awaiting recovery, and waste awaiting disposal. If this control is to be implemented effectively and economically, nondestructive assay techniques must play a central role. These same nondestructive assay techniques when further refined can also play an important role in improved quality control --an important step towards universal acceptance of nuclear power.

At Los Alamos, research in nondestructive assay of fissionable materials in support of nuclear safeguards has been in progress since 1967.[2] The program includes assay by gamma counting, by counting spontaneous-fission or (α, n) neutrons, and by the detection of neutron-induced fission. It is the last method of assay to which the remainder of this paper is directed.

Inventory Verification Samples

As a part of the program of safeguards inventory control, samples of material are withdrawn by AEC inspectors from production lines and from containers in storage vaults. These samples are now sent to the AEC New Brunswick Laboratory for fissionable assay by chemical analysis at a cost of from near $50 to well over $200 each, depending upon the chemical nature and radiological properties of the material. The requirements for inventory verification, with stricter controls than heretofore in a rapidly growing industry, could well lead to an order-of-magnitude increase in the number of inventory verification samples--currently numbering about 5,000--which must be analyzed each year. Nondestructive assay of these samples offers the possibility of an order-of-magnitude saving in the annual cost of the operation. The small-sample assay problem has been pursued along numerous channels including interrogation with pulses of unmoderated and moderated 14-MeV neutrons followed by delayed-neutron counting,[2] counting of high-energy prompt neutrons from fission while interrogating with 25- to 35-keV ^{124}Sb-Be neutrons,[3] and use of thermal-neutron interrogating systems using ^{252}Cf.[4] Passive assay by gamma-ray counting with suitable corrections for self-absorption[5] has also been applied to this problem.

Assay by Delayed-Neutron Counting

Our most effective method for assay of ^{235}U in nonhydrogenous inventory samples is based upon interrogation of the samples with 300- to 600-keV neutrons, produced by bombarding 2-mg/cm^2-thick lithium targets with 2.35-MeV protons from the LASL 3.75-MeV Van de Graaff accelerator. The modulated source technique developed by Masters, Thorpe, and Smith[6] is used to measure the delayed-neutron yield from the samples. Briefly this technique 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[6] that if the neutron bombarding cycle is operated for a time long with respect to the longest half-life of the delayed-neutron groups (55 sec), then the delayed-neutron counting rate 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. For relative delayed-neutron yield measurements used in assay, however, it is not necessary to wait for secular equilibrium of the delayed-neutron groups; it is only necessary to start the count of all samples and standards either at the beginning of the irradiation or at some fixed time (we use 30 to 50 seconds) from the start of irradiation.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

Fission chambers are used to monitor the interrogating flux. These should be in the same neutron environment as the sample, so that one may at least partially compensate for flux-depression effects. The fission chambers should be 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, it can be shown 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 about 6 mg of ^{235}U for our best available source strength of about 10^{11} neutrons/second. Beam-induced backgrounds are equivalent to about 2 mg of ^{235}U (the content of the fission chambers!), indicating that we are achieving on-to-off ratios in excess of 10^9.

Facility Requirements

Achievement of a high neutron 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 1 shows the layout of the LASL 3.75-MeV accelerator facility. Three feet of concrete shield the experimental area from the accelerator. 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 100-μA beam prior to acceleration. For this reason, beam pulsing is accomplished in the beam line after acceleration and momentum analysis, by means of a rotating mechanical shutter and synchronized electrostatic deflector. The mechanical shutter is shown in Fig. 2. Figure 3 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.

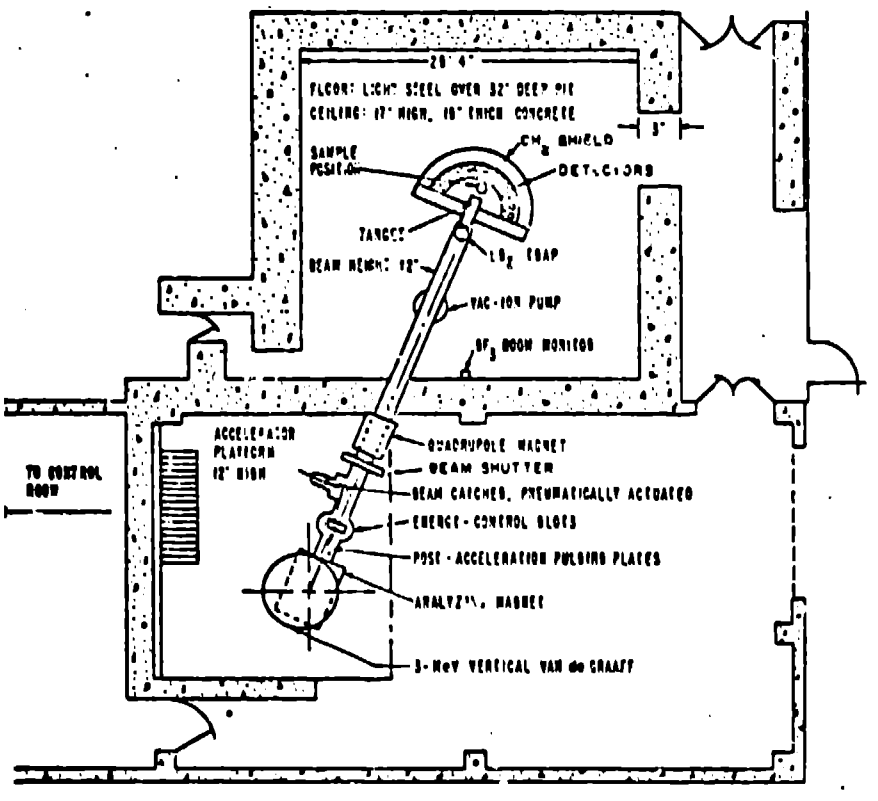


Fig. 1. Layout of LASL 3.75-MeV Van de Graaff accelerator facility.

Fig. 2. Rotating mechanical beam shutter.

Fig. 3. View of accelerator horizontal beam line, showing electrostatic deflector, control slits, water-cooled pneumatic beam-catcher/stop, and neutron shielding about mass-2 port of the momentum-analyzing magnet.

Effect of Low-Energy Neutrons

We initially tried small-sample assay by measurement of delayed-neutron yields using the geometry shown in Fig. 4. The delayed-neutron detector was the "slab detector" of Fast and Walton,[7] 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 1/4-in. Boral plate between the slab detector and the sample noticeably reduced matrix dependence. This is shown in Fig. 5. 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.

The matrix-dependence effects were due to the presence of epicadmium, resonance-region neutrons which caused the samples to exhibit large self-absorption effects. To eliminate this problem, a small-sample assay chamber was built with 1.9-cm-thick hot-pressed B_4C walls. Using the B_4C shield, which is is calculated to have an effective neutron cutoff of about 100 eV, the bottom curve is obtained. Another set of tests was performed using various amounts of pure enriched U_3O_8. This is shown in Fig. 6.

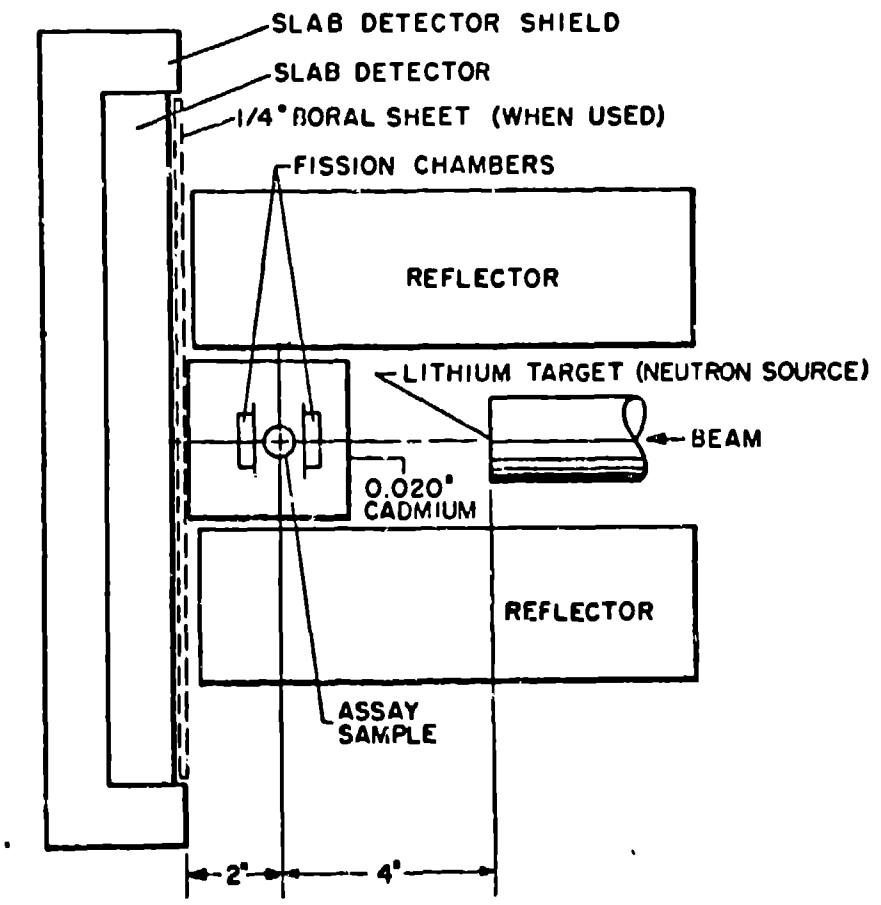


Fig. 4. Layout for early efforts to assay small samples.

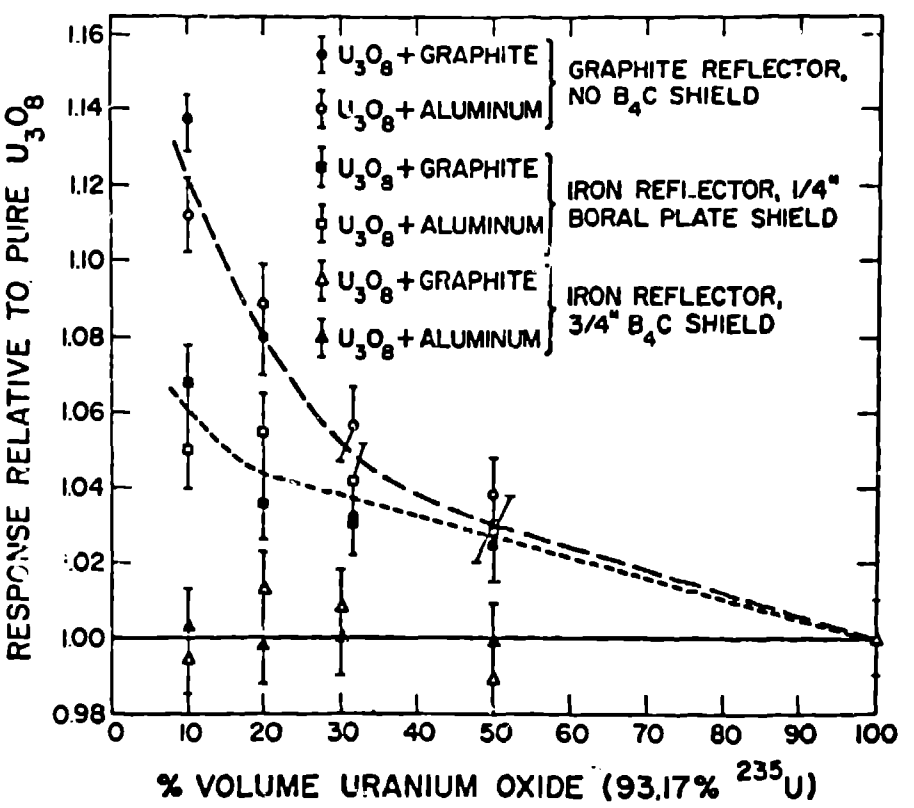


Fig. 5. Comparison of the effect of dilution of 1 g of enriched uranium with graphite or aluminum upon the delayed-neutron assay response. Upper curve: Cd thermal shield only. Middle curve: partial epithermal shield of 1/4-in. Boral plate. Lower curve: assay using a 3/4-in. pressed boron carbide shield about sample and fission chambers.

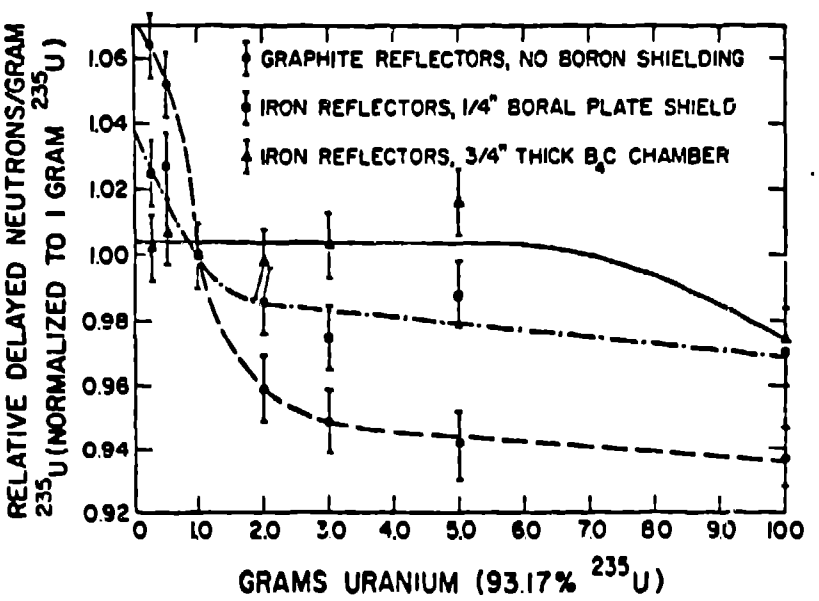


Fig. 6. Effect of epithermal shielding upon the linearity of delayed-neutron assay response to pure U_3O_8 standards.

Application

Experience with the assay of one type of fuel ıaterial demonstrates the utility of the method. High ɛmperature gas-cooled reactors are fueled with eleıents containing silicon-carbide coated microspheres f mixed enriched uranium carbide and thorium carıide. This material is intended to be highly resistant ɔ chemical attack--and it is. As a result, chemical ssay costs from $200 to $400 per sample of a few cm^3 ontaining approximately 250 mg of ^{235}U and several rams of thorium. Because of dissolution problems, he results of chemical assay have a precision at preent of only about 1% at the 1σ level. These beads can e assayed to the same precision by neutron interrogaion and delayed-neutron counting with a throughput of 0 samples per day. If one considers $50/h a reasonble estimate of the cost of operating a small accelertor, the cost of an assay comes out to a little less han $10. The results of a recent exercise involving his material are shown in Fig. 7. Samples of the mirobeads were assayed using as a reference standard . vial containing 0.6 g of highly enriched uranium as U_3O_8, 6.0 g of Th as ThO_2, and 7.75 g of powdered graphite. An effectively zero bias between chemical nd nondestructive assay is testimony to the indepenlence of the nondestructive assay results in this case o the chemical and physical form of the material. It hould be noted that, if thermal neutrons had been used or the assay, the response would have been greater nd the resultant statistical precision better, but selfbsorption in individual fuel beads would have resulted n a negative assay bias of several percent.

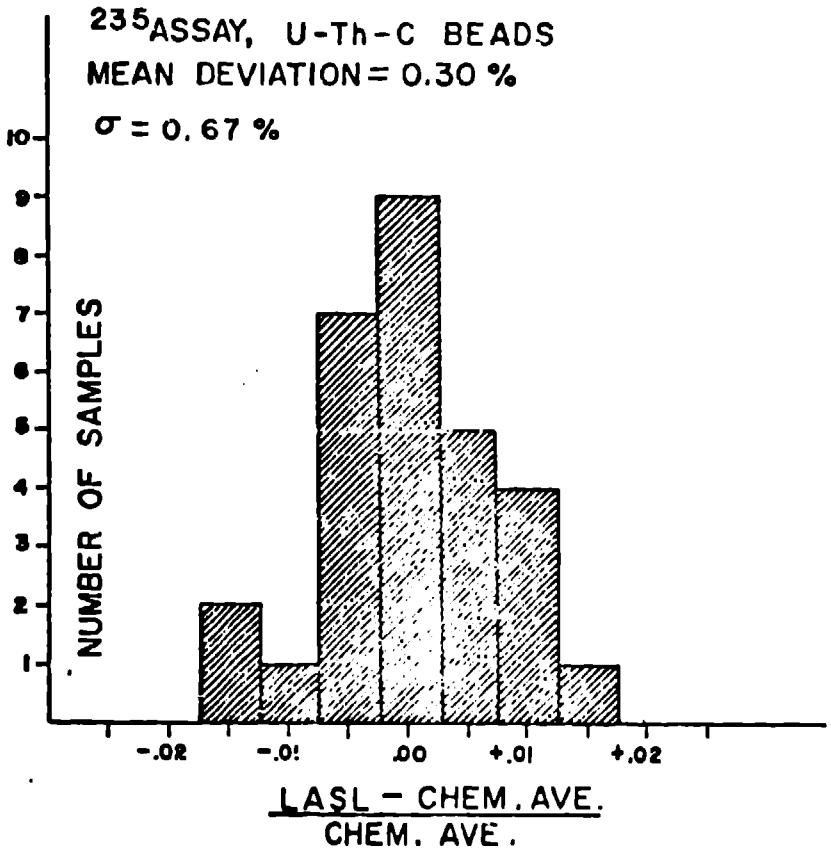


Fig. 7. Test of small-sample precision and accuracy using 30 vials (presumed identical) of HTGR U-Th-C microspheres. Results are normalized to the average chemical analysis of the vial contents.

Production Assay System

An automated assay system has been built to put to work the principles outlined above. The heart of this system, shown in Fig. 8, is an efficient neutron counter assembly, the active portion of which is a 41-cm i.d. by 15.2-cm-thick by 61-cm-high semicircular polyethylene slab containing 39 2.5-cm-diam by 50-cm-long 4-atm ^3He proportional-counter tubes. The detector tubes are connected in two banks, detectors closest to the inside wall in one bank and those close to the outside wall in the rear bank. With this arrangement the front-to-rear bank counting ratio is a measure of the average energy of incident neutrons; however, the rear counter bank of the detector was placed somewhat forward of the optimum "flat response" position used in the design of the slab detector of East and Walton[7] in order to increase the overall counting efficiency.

The detector is completely surrounded by a 15-cm-thick boron-lined polyethylene shield through which the Van de Graaff beam line penetrates to provide a neutron source 10 cm from the sample position. The sample to be assayed is surrounded, together with the monitoring fission chambers, by 1.9 cm of pressed boron carbide and an inner sleeve of 0.5 mm of cadmium. Sliding front shield doors provide access to the accelerator target and the sample irradiation area. A 5-cm-thick nickel reflector inlaid into the shield doors behind the accelerator target and a nickel collar extending from the shield doors to the plane of the accelerator target increase the interrogating neutron flux at the sample by 50% and also serve to reduce spatial dependence of the sample response by increasing the effective size of the neutron source. A pneumatic transfer system is used to change samples.

Fig. 8. "Super Slab" small-sample assay system. A: ^3He-tube-containing sensitive volume; B: detector-tube junction box; C: fixed polyethylene shielding; D: movable polyethylene shield doors; E: pressed B_4C sample-irradiation chamber; F: pneumatic sample tube; and G: accelerator target (neutron source).

Figure 9 is a view of the assay station with one shield door and most of the B_4C sample shield removed to show construction of the detector and the location of the flux-monitoring fission chambers.

With this system, a sample containing 250 mg of ^{235}U can be assayed to a precision of 1% at the 1σ level in 400 sec of counting time. With backgrounds at their present levels, the capability exists to assay as little as 5 mg of ^{235}U or the fissile content of ~1 g of natural U. About 45 sec are used between runs to change samples and allow 30 sec for buildup of the delayed-neutron activity. Data from scalers are recorded on paper tape for analysis on a PDP-9 computer system. One man operates the assay system and the Van de Graaff accelerator.

Tests are now underway to determine the ultimate capability of the new system. It is believed that this system will prove to be a practical means for reducing the cost of analysis of inventory verification samples and will also play a role in the more exacting field of quality control. One important role for which the system appears suited is the intercomparison of small-sample nondestructive assay standards, a facility need which is growing rapidly.[8]

Fig. 9. "Super Slab" with one shield door and B_4C sample chamber removed. A: sensitive volume; B: detector-tube junction box; C: fixed polyethylene shielding; D: movable polyethylene shield door; E: flux-monitoring fission chambers.

References

1. U. S. Code of Federal Regulations 10CFR70; Federal Register 38, No. 21 (1973), p. 3077.

2. G. R. Keepin, R. H. Augustson, and R. B. Walton, Proceedings of the AEC Symposium on Safeguards Research and Development, Los Alamos Scientific Laboratory, October 27-29, 1969, (WASH-1147, 1970), p. 109.

3. H. O. Menlove and R. A. Forster, Los Alamos Scientific Laboratory report LA-4994-PR, p. 6 (1972).

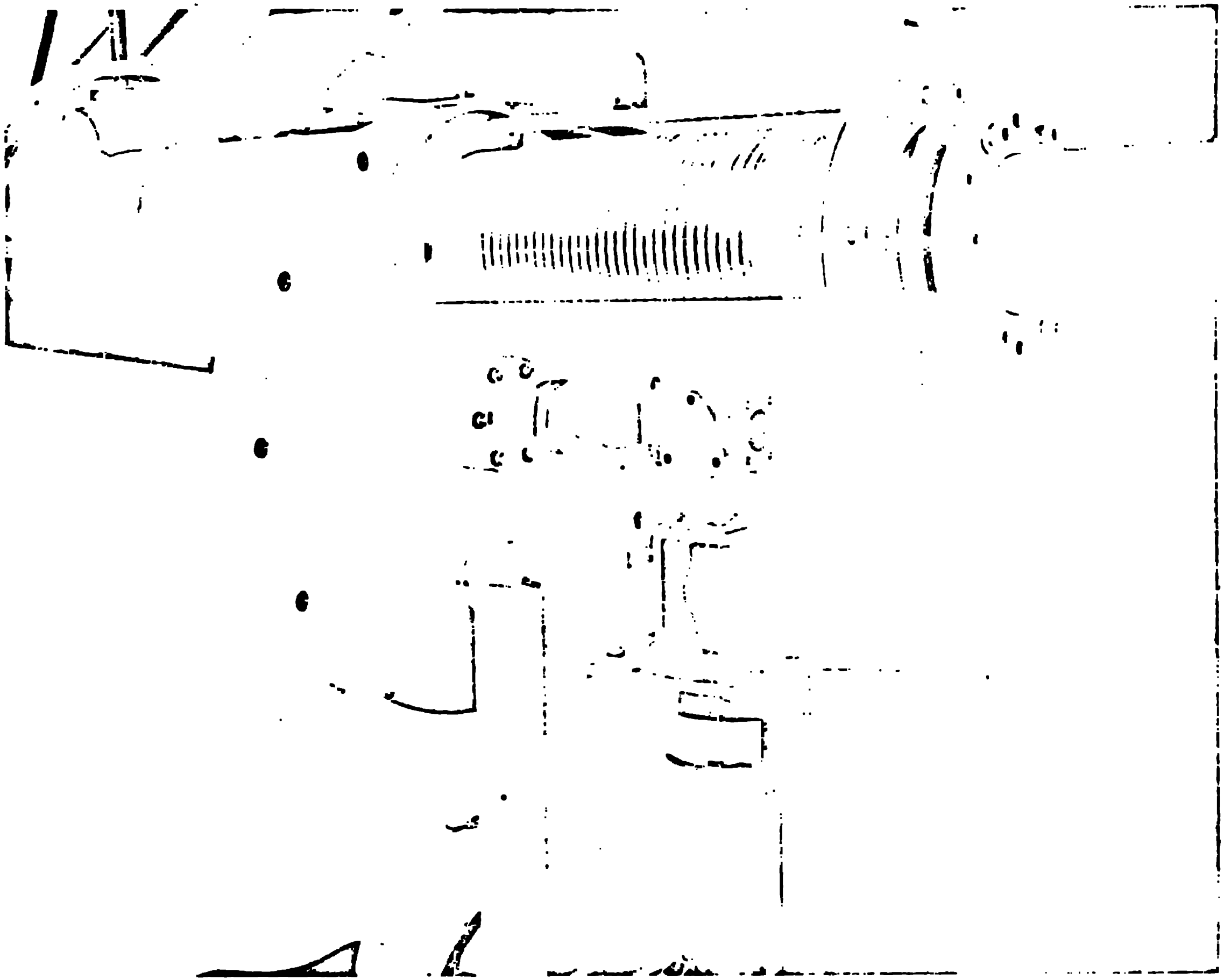
4. H. O. Menlove, R. A. Forster, and J. L. Parker, Los Alamos Scientific Laboratory report LA-4994-PR, p. 5 (1972).

5. J. L. Parker, T. D. Reilly, R. B. Walton, D. B. Smith, and L. V. East, Los Alamos Scientific Laboratory report LA-4705-MS, p. 12 (1971).

6. C. F. Masters, M. M. Thorpe, and D. B. Smith, Nuclear Science and Engineering 36, 202 (1969).

7. L. V. East and R. B. Walton, Nuclear Instr. and Meth. 72, 161 (1969).

8. D. M. Bishop, "Nondestructive Assay Measurement Standards: Need and Approach," Journal Institute of Nuclear Materials Management II, No. 3 (1973).



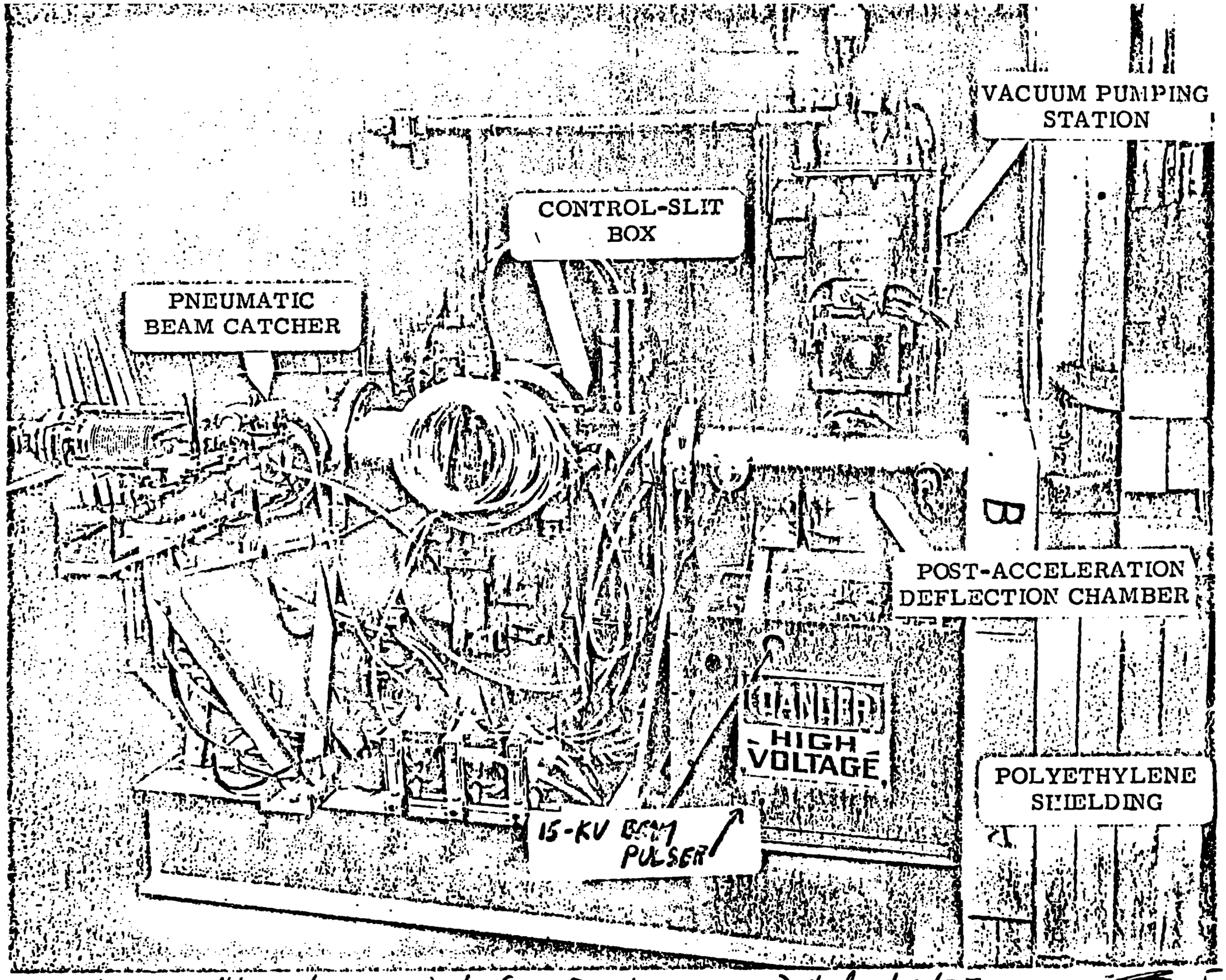
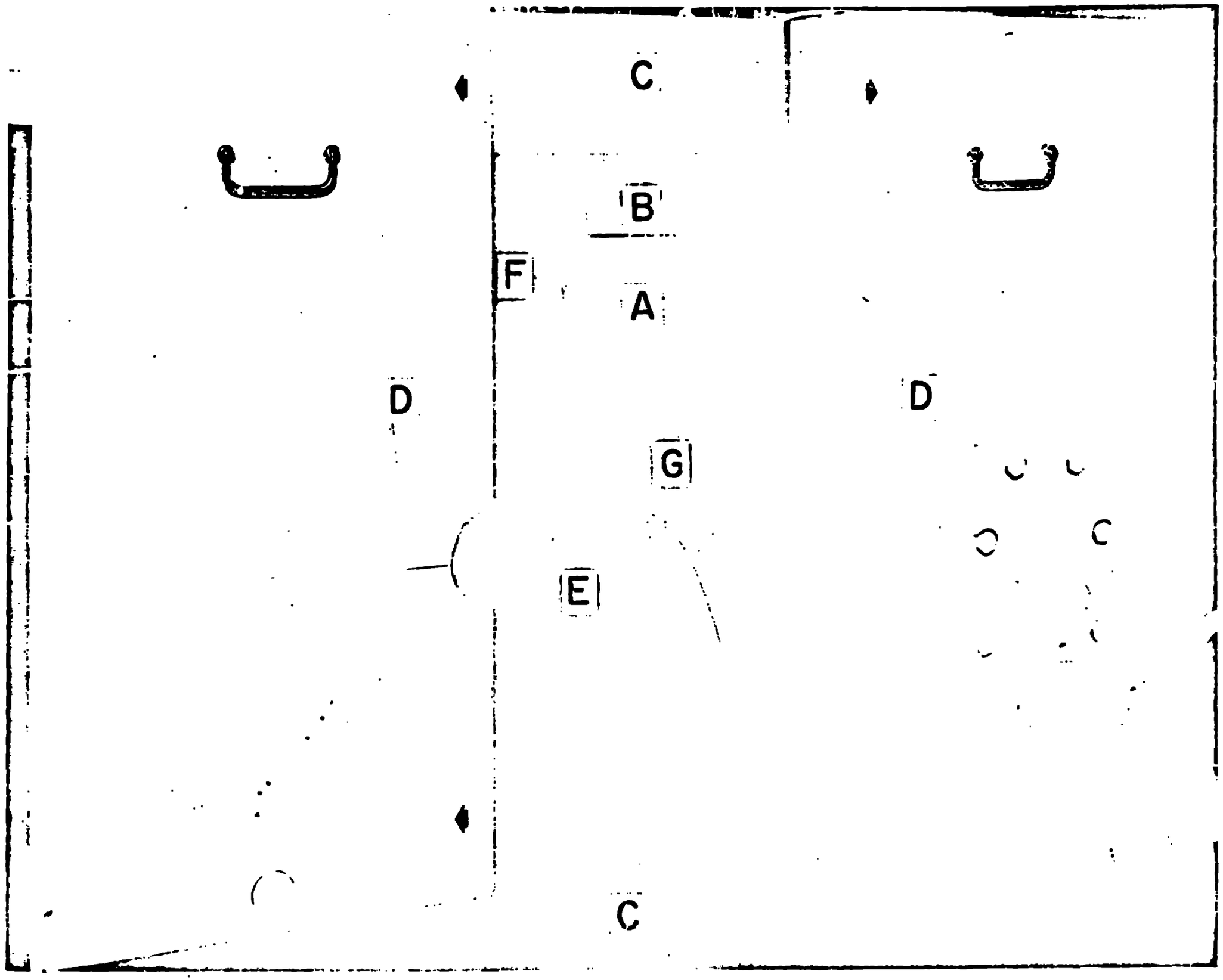

VACUUM PUMPING STATION
CONTROL-SLIT BOX
PNEUMATIC BEAM CATCHER
POST-ACCELERATION DEFLECTION CHAMBER
DANGER HIGH VOLTAGE
POLYETHYLENE SHIELDING
15-KV BEAM PULSER

Fig 3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