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THE ROLE OF NUCLEAR DATA IN THE PRACTICAL APPLICATION OF NONDESTRUCTIVE NUCLEAR ASSAY METHODS*

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INTRODUCTION

Practical nuclear nondestructive assay methods for fissionable material gaining acceptance today are based mainly on two fundamental signatures. First, the naturally occurring radioactivity characteristic of the nuclides of interest and second, the ability of these same nuclides to be fissioned either by neutrons or gamma rays. With some important exceptions, the basic nuclear data relating to these properties has been available for a number of years. Why then are nuclear data important? The answer lies in the diversity of purpose of the assay methods. The hundred-thousand-dollar machine designed to provide precise assay of several million dollars worth of fuel pins is hardly comparable to the device used for the occasional measurement of a dozen or so waste barrels. Because of this diversity of purpose an array of instrumentation is being developed, each making use of the basic signatures in a different way to satisfy a different set of conditions.

Uses of nuclear data in the development of practical nuclear assay techniques fall naturally into three categories. First, there are parameters which can enter directly into the calibration of equipment, such as decay rates, gamma intensities and attenuation coefficients, neutron yields, etc. The second category involves the use of nuclear data in the predictive sense as input to neutron and gamma-ray transport codes which provide calculational support and guidance to the safeguards research and development program. This support is necessary for the detailed understanding of tech-

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niques under investigation as well as for design optimization of systems already proven to be worthy of development to the prototype stage. The third category utilizes nuclear data as a reservoir of basic information to aid in the search for new methods and signatures.

FUEL PIN ASSAY

A number of fuel pin assay systems either complete or under development serve as an illustration of some of these uses of nuclear data. These systems are for the measurement of:

1. Uranium-235 content of LWR fuel.
2. Uranium-235 content of LWR fuel and the detection of out of specification fuel pellets.
3. Plutonium content and isotopic composition of fast breeder reactor (FBR) fuel.
4. Plutonium recycle LWR fuel.
5. Fissile content of irradiated fuel.
6. Uranium-235 content of LWR fuel which also contains burnable poison.

Three properties of the system for measuring LWR pins are most important:

1. High throughput rate to take care of a modern plant capacity of several hundred pins per shift.
2. Sensitivity to all of the fissile material within the pins.
3. Reliability.

These three criteria lead to the choice of thermal neutron interrogation; the thermal flux supplied by a moderated ^{252}Cf source, and the detection of prompt fission neutrons from the thermal fission of ^{235}U furnishing the primary signature and differentiation from ^{238}U, the major constituent of LWR fuel.

The moderator for the neutron source must provide sufficient thermal flux at the fuel channel while providing maximum discrimination between ^{235}U and ^{238}U as well as minimizing the source strength and biological shielding required. The configuration of source, moderator materials and detectors was optimized by modeling the system on the computer using only enough actual experimentation to assure confidence in the computer calculations [1]. The method of optimizing a design by computer calculations illustrates one of the more important uses of nuclear data as input to computer codes which provide the calculational tool necessary to design practical nondestructive assay instrumentation.

The assay of a fuel pin requires a mathematical calibration function which describes as accurately as possible the response for various enrichments and yet is simple enough to permit on-line data reduction. Analysis [2] indicates that the response of a fuel rod having an enrichment less than 4% ^{235}U is adequately described by a function of the form

$$R = A(1 - e^{-BU}) \quad .$$

R is the measured response, U is the ^{235}U content of the fuel rod, and A and B are parameters determined from measurements of standard fuel pins.

In addition to the ^{235}U content, the response of an actual fuel pin depends on a number of factors: cladding thickness and composition, fuel section length, density, diameter, impurity content, etc. It is important to investigate the sensitivity of the assay system to changes in these various parameters. Here again, the computer and nuclear data are used to obtain the information. These calculations and other considerations provide the basis for a systematic error analysis [3]. The precision of a single fuel rod assay is dominated by propagated counting statistics amounting to ~1.1 to 1.5% (1σ) over the mass range of 30 to 120 g of ^{235}U per rod. After 100 fuel rod assays, the statistical uncertainty in the total mass decreases to less than 0.15%. At this point the error becomes dominated by factors other than counting statistics (e.g., the error in the calibration curve).

In-plant experience has proven the practicality of measuring the entire ^{235}U throughput of a modern plant. The system can be operated by plant personnel with little technical training at a rate of 400- to 600-rod assays per eight-hour shift. This rate includes the time taken for pin identification, handling, and the assay of several standard rods to provide a calibration check. A more detailed description of the construction and operation of the units may be obtained from a paper by R. A. Forster, et al. [4].

A series of Monte Carlo calculations and measurements [5] led to the following modifications of the thermal neutron ^{252}Cf fuel rod system to include measurements of pellet-to-pellet variations: 1) lengthening the moderator assembly in the direction of fuel rod travel, which results in a longer region of high thermal flux; 2) positioning the fuel rod channel closer to the ^{252}Cf source to take advantage of the higher intensity flux near the source while still preserving a high fissile/fertile fission ratio (~10^5) to ensure accurate measurement of fissile content in the presence of much larger amounts of fertile material; 3) replacing the D_2O in the moderator assembly with deuterated water-extended-polyester resin and carbon to decrease cost while simplifying the fabrication and shipping; 4) changing the fuel channel position requires that the ^3He detectors be shortened and displaced to one end of the carbon moderator to give a sufficiently large signal/^{252}Cf background ratio in the ^3He detectors; and 5) the addition of a small (2 by 2 by 3/4 in.) NaI detector near the exit port of each fuel-rod channel to give pellet-to-pellet fissile content by counting fission-induced gamma rays. Figure 1 is a schematic diagram of the Pin and Pellet Assay System (PAPAS). Figure 2 illustrates the system capability for the detection of off-specification pellets.

A complete assay and quality control station for FBR fuel rods has been delivered to Westinghouse-Hanford Engineering Development Laboratory (HEDL). These rods contain mixed oxide fuel with a $^{239}Pu/^{238}U$ ratio of ~1/4, and a total ^{239}Pu mass of ~30 g. The instrumentation has been calibrated and is now in routine operation. The station consists of two units. One unit [6] contains a ^{252}Cf (619 μg) source tailored to provide fast neutron irradiation of the pins. The delayed gamma rays from fission (E_γ > 1.2 MeV) are detected as a measure of the total fissile content. The gamma rays from ^{241}Am, ^{239}Pu are counted and used to provide pellet-to-pellet information. The second unit measures the ^{240}Pu content by coincidence counting the neutrons emitted from the spontaneous fission of ^{240}Pu [7].

Because of the high fissile loading of FBR fuel pins, thermal neutron irradiation would have yielded an unacceptably nonlinear response curve. Calculations led to the choice of the cylindrical moderator assembly shown in Fig. 3. The assembly has a core of tungsten (2.5 cm radius) surrounded by a 7.5-cm-thick shell of beryllium followed by 5 cm of lead and nickel. The nickel reflector increased the fission rate ~70% over the lead reflector alone. This moderator design resulted in a $^{239}Pu/^{238}U$ fission ratio of ~400/1 for irradiation neutrons above the cadmium cut-off energy (~0.4 eV).

Figure 4 is a schematic diagram of the unit showing the NaI detectors each having a different degree of collimation, moderator, and shield assembly. Americium-241 doped, NaI seeds embedded in each crystal provide a source of constant amplitude pulses (roughly equivalent to a 3 MeV gamma ray) which are used in conjunction with an electronic stabilizing unit to reduce the effects of long-term photomultiplier and electronic drifts. Figure 5 is a photograph of the entire unit showing the moderator-shield assembly, the pin handling equipment, the programmable calculator and associated electronics. The automated translator picks up the rod to be assayed and moves it through the NaI crystals and the ^{252}Cf assembly at a rate of 2.5 in./sec in order to take a background count of the unirradiated rod. The direction of travel is then reversed and the rod is withdrawn at 0.36 in./sec during which time the delayed fission gamma-ray data are acquired and the pellet-to-pellet scans are made. The rod is then unloaded in the tray directly below the loading magazine and the cycle is repeated for the next rod. The translator speeds are set to give a 5-min cycle per rod.

Error analysis and measurements indicates that the standard deviation of the measured fissile content of a rod is ~0.2 g. Figure 6 illustrates the sensitivity to pellet-to-pellet changes as observed with a test pin containing various pellet and enrichment combinations. It can be seen that small changes in enrichment (1.7% to 3.1% relative) can be detected. The ^{240}Pu assay system, shown in Fig. 7, contains 32 ^{3}He tubes (1 in. diam by 20 in. long, 4 atm gas pressure). The detector efficiency is ~36% and the neutron lifetime was measured to be 28 μsec. Assay precision is a little greater than 1% (1σ) for a counting time of 100 sec.

Assay units using Sb-Be or ^{88}Y-Be as fast, subthreshold neutron sources are under investigation. A relatively hard nonthermal interrogation flux offers the potential advantage of insensitivity to the presence of fission product or burnable poisons as well as providing good penetrability and linearity of response. Figure 8 is a photograph of an experimental photoneutron system [8]. There is a central cadmium lined assay channel in beryllium with adjacent holes for the gamma sources followed by rings of titanium and nickel. Three inches of lead shield the ^{3}He detectors from the source gamma rays. The ^{3}He detectors are embedded in a thick (2 cm) nickel ring which serves as a fast neutron reflector. The nickel reflector added about 40% to the signal rate. The energy of the neutrons from the photoneutron sources is such that the ^{3}He detectors can be biased to eliminate most of the neutron signal from the source. This in turn permits the addition of sufficient lead to shield the detectors from the intense gamma radiation of either the source or the sample. The photoneutron unit just described is being used to acquire data on optimum detector gas pressure, signal rates and signal-to-background ratios, response linearity, etc. The unit is also being used to determine its suitability for other applications such as small-sample assay [9].

LWR fuel containing recycled plutonium will become important in the near future. The special assay problems associated with this type of fuel are being studied.

NEUTRON METHODS APPLIED TO SCRAP AND SMALL SAMPLE ASSAY

One of the most significant problems associated with the assay of scrap and waste is the lack of control over the extraneous material that might be present within the sample. Assay by means of fast neutron irradiation and detection of delayed or prompt fission neutrons has been found to be relatively insensitive to all matrix materials except hydrogen and other low Z elements which are good neutron moderators. Fission chambers (or other detectors sensitive to low energy neutrons) closely coupled to the sample provide a means of detecting or correcting for the effects of neutron moderations. Gamma-ray assay methods are most suitable for light, hydrogenous matrix materials. Together, the two techniques, gamma-ray and neutron assay methods, provide broad capability for many of the assay problems associated with scrap and waste.

The development of the technique of neutron interrogation with delayed neutron detection is an example of the key role that nuclear data can play. When this method was first being considered it was necessary to undertake an experimental program to measure the delayed neutron yield from fission as a function of incident neutron energy. Initial emphasis was placed on measurements at 14 MeV [10] since 14 MeV neutron generators are a relatively inexpensive, copious source of neutrons. This program was followed by another set of measurements using a Van de Graaff as a variable energy neutron source [11].

These experiments were designed to confirm and extend the available delayed neutron yield data. The results show that the delayed neutron yield is not significantly dependent on the energy of neutrons causing fission for energies below 5 MeV. Above 5 MeV there is a drop in yield corresponding to the onset of second chance fission. The data obtained so far are in general sufficient to satisfy most immediate practical needs. Nonetheless there is interest in the delayed neutron yield in the energy region 6 to 14 MeV and in data for the higher plutonium isotopes. These basic data are used for computer calculations to explore and define possible technique refinements.

An example of the application of subthreshold neutron interrogation and delayed neutron detection is the fissile assay of small samples [12] taken from various portions of a plant inventory for process control or inventory verification. Several thousand such samples are chemically analyzed yearly. To be generally useful, a small sample assay technique must be able to furnish few percent or less accuracy for a wide variety of chemical forms and concentrations of fissile material. Figure 9 is a photograph of a small sample assay apparatus which utilizes a Van de Graaff accelerator as a pulsed source of few hundred kilovolt neutrons. The delayed neutrons are detected between accelerator pulses by a large, flat efficiency detector. Closely coupled fission chambers monitor the fission rate in the samples. A background equivalent of 15 mg ^{235}U has been achieved.

Assay precision depends on sample fission rate and detector efficiency. It is advantageous to closely couple the detector and to provide neutron reflectors to increase the flux at the sample. The presence of extraneous

materials in the vicinity of the sample degrades the incident neutron energy and causes undesirable sample self-absorption and matrix effects. An acceptable compromise, found experimentally, is to use iron reflectors adjacent to the target and sample in conjunction with a 3/4-in.-thick boron carbide sleeve surrounding the sample to eliminate the majority of neutrons below 100 eV. Favorable experience gained to date from several hundred assays has provided the incentive to undertake the following major improvements in the system: automatic sample handling, automated data processing, and a more efficient detector-reflector geometry. Ongoing research effort involves finding methods of increasing precision, reducing the number of standards required, and defining and eliminating sources of bias.

GAMMA METHODS

Gamma-ray spectroscopy, particularly with lithium drifted germanium (GeLi) detectors, is a general purpose method with a wide range of applications. In this instance, nuclear data is not so important in the design of the hardware, but is essential for its application. Usually only a few lines from the complex decay spectra of the nuclides of interest are used for assay. Nonetheless extensive knowledge of gamma spectra is required to provide the assurance that the lines used are specific and that the presence of unusual or unsuspected activity will not yield an erroneous assay. Attenuation corrections for the matrix materials involved are the major sources of uncertainty in the assay. Attenuation corrections are usually obtained through: preparation of standards which are representative of the material being measured; measurement of the transmission of a source [13]; comparison of the relative intensities of two or more characteristic gamma rays [14].

In addition to quantitative assay gamma-ray detection has been applied to the measurement of enrichment, concentration, and isotopic composition. Relatively simple instrumentation, particularly when used by knowledgeable personnel, can be quite effective. Figure 10 shows one of the devices which was used to estimate the holdup in a shutdown diffusion plant cascade. The instruments were also used in the operating cascade to monitor UF_6 retention in NaF traps, measure enrichment and to detect plating or holdup in the main gas pipes [15].

COINCIDENCE COUNTING OF FISSION EVENTS

Coincidence detection of the many neutrons and gamma rays from fission provides a convenient method for separating the occurrence of fission from the source producing the fission or from extraneous radiation which might be present [e.g., neutrons from (α,n) reactions]. Neutron coincidence counting of ^{240}Pu spontaneous fission provides a simple method of plutonium assay when the isotopic composition is known. The spontaneous fission rate of ^{238}U can also be used for assay purposes. Both rates are low and high efficiency 4π neutron counters are required for rapid quantitative assay.

Organic scintillation detectors permit the detection of time correlated events, neutrons or gamma rays, using coincidence gate widths of only a few tens of nanoseconds. Since the ratio of accidental coincidence rate to true coincidence rate is directly proportional to the coincidence gate width, the short gate width allows the detection of fission in the presence of relatively large backgrounds of uncorrelated neutrons and gamma rays. Conversely, an uncorrelated or random source can be introduced to cause fission in the sample.

The detected fission rate then is a measure of the fissionable content. Neutron source energies can be changed to increase sensitivity, and to provide both fissile and fertile assay. Figure 11 is a photograph of a system called the "Random Driver" which uses an Am-Li neutron source to cause fission in the sample. This system proved an effective instrument for the assay of uranium [16,17].

BASIC SIGNATURES

Although natural radioactivity and fission have furnished the basic signatures which have proven to be the most utilitarian, there are other characteristics which can be used to identify particular elements or isotopes. A few examples are: neutron capture gamma rays; delayed-neutron and gamma-ray spectra, gamma-ray and x-ray fluorescence; selective neutron and gamma-ray absorption. The gathering of information, particularly basic data, pertaining to any physical phenomenon which might be applicable to materials analysis should be encouraged. This reservoir of basic information can then be used as a basis to continually review the techniques for possible application to the changing and differing needs for quantitative assay. These needs range from the detection of trace quantities in effluents to detailed analysis of spent reactor cores. New facilities and improvements in detector characteristics, for example, may render completely practical a technique previously thought not worthy of further development.

An example of the information gathering process is a program that is designed to investigate the application of μ-meson capture x-rays to elemental and isotopic analysis. The objective of this program is to obtain high quality spectra for each fissionable isotope and to obtain information on how the chemical form of the material might affect possible assay applications. Using the facilities at the Space Radiation Effects Laboratory (NASA) data have been taken on metal targets of ^{208}Pb, ^{232}Th, ^{235}U, ^{238}U, and ^{239}Pu. Some data were also obtained for depleted uranium compounds, mainly oxides. The experiments are expected to continue early next year when more intense beams become available at the Los Alamos Meson Physics Facility.

CONCLUSIONS

The number of systems for assay that exist, complete with operations manual, error analysis, and operational history of reliability and effectiveness, is testimony to the growing maturity of nondestructive assay. A more difficult phase is beginning which emphasizes accuracy without undue increase in cost and complexity, the development of standard procedures, and the establishment of nondestructive assay methods as independent alternatives to traditional chemical analysis.

Calorimetry is an example of a technique for which refined nuclear data would have a direct effect on meeting the challenges listed above. The radioactive decay data and methods of determining isotopic composition are not sufficiently accurate to establish the relationship of heat output and quantity of material to an accuracy comparable to the precision available [18].

The value of readily available data which form the basis for design calculations and the foundations from which to explore new concepts can hardly be overemphasized. Routine calibration of equipment is accomplished by means of standards. Improvements of the data and calculational techniques will permit more precise evaluation of system performance which will reduce the number of these costly standards required.

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LIST OF FIGURES

1. Thermal neutron ^{252}Cf fuel-rod assay system with modifications for pellet-to-pellet scan. The ^{4}He neutron detectors in the carbon core count the prompt fission neutrons for total fissile determination, and the NaI detectors near the fuel-rod exit channel count the delayed gamma rays for pellet-to-pellet determination.

2. A typical delayed gamma-ray scan of a 66-in.-long 3.3% PWR fuel rod with pellets of lower enrichments interspersed as shown above. The lower curve is a smoothed version of the raw data in the upper curve (the error bars represent 2σ uncertainties). Each point represents the total counts accumulated in 0.4 sec for a rod feed rate of 8 ft/min.

3. Photograph of moderator and shield assembly for a fast neutron ^{252}Cf assay system. The ^{252}Cf source is positioned in the center of the tungsten and the sample is placed in the nickel reflector for the neutron irradiation.

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6. FFTF fuel pin with various combinations of plutonium enrichments for pellet-to-pellet scanning. Top curve corresponds to 60 keV energy window and bottom curve corresponds to 100 to 500 keV window.

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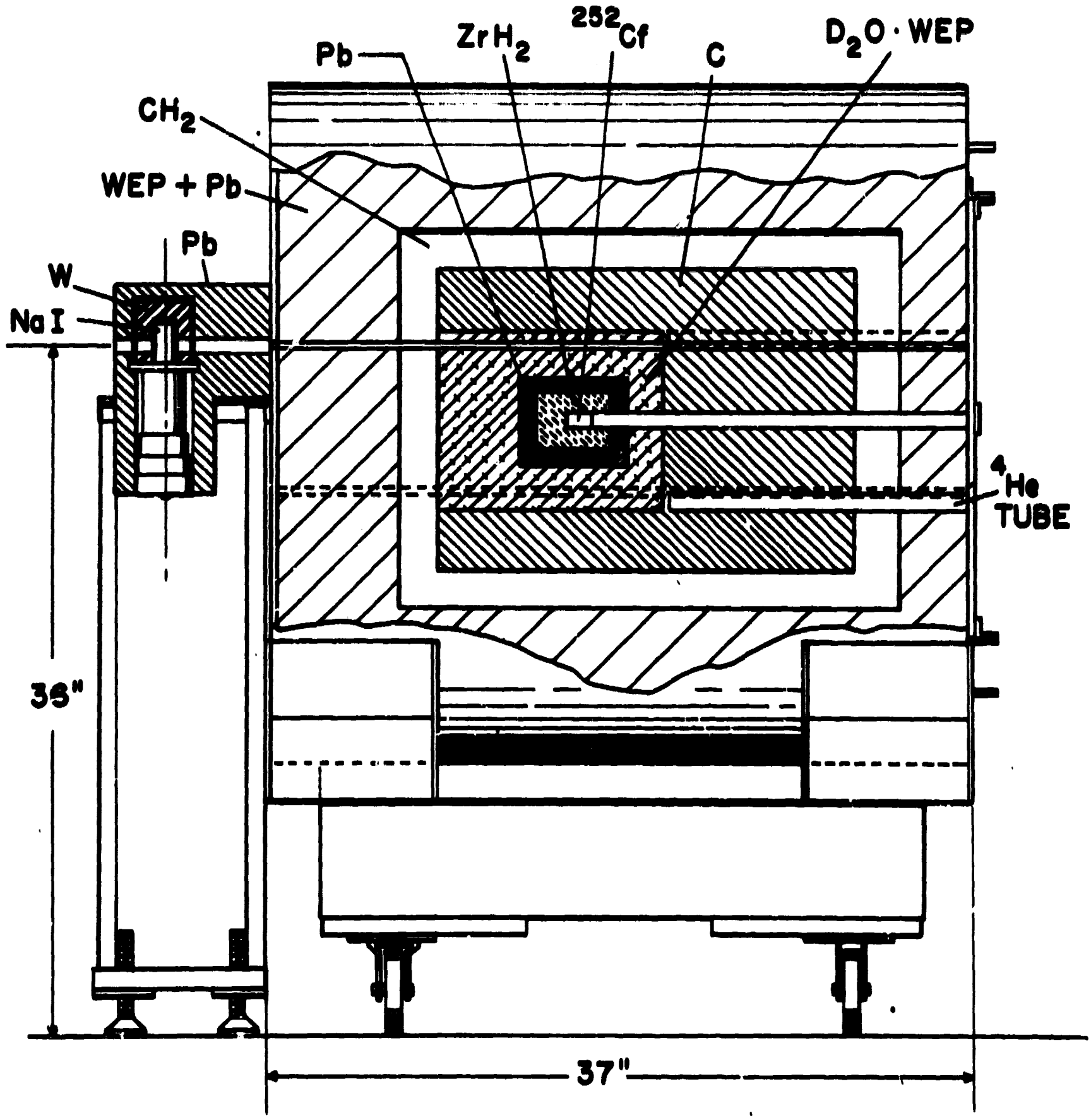
8. Photoneutron assay system using either ^{124}Sb or ^{88}Y in the beryllium core surrounded by nickel and titanium neutron reflectors and lead gamma-ray shielding.

9. View of small-sample assay station with B_4C shield removed.

10. Cooled, portable NaI gamma spectrometer for assay of ^{235}U inside an operating gaseous-diffusion plant.

11. The random source-interrogation system used to determine the ^{235}U content in containers of up to 5-gal capacity.



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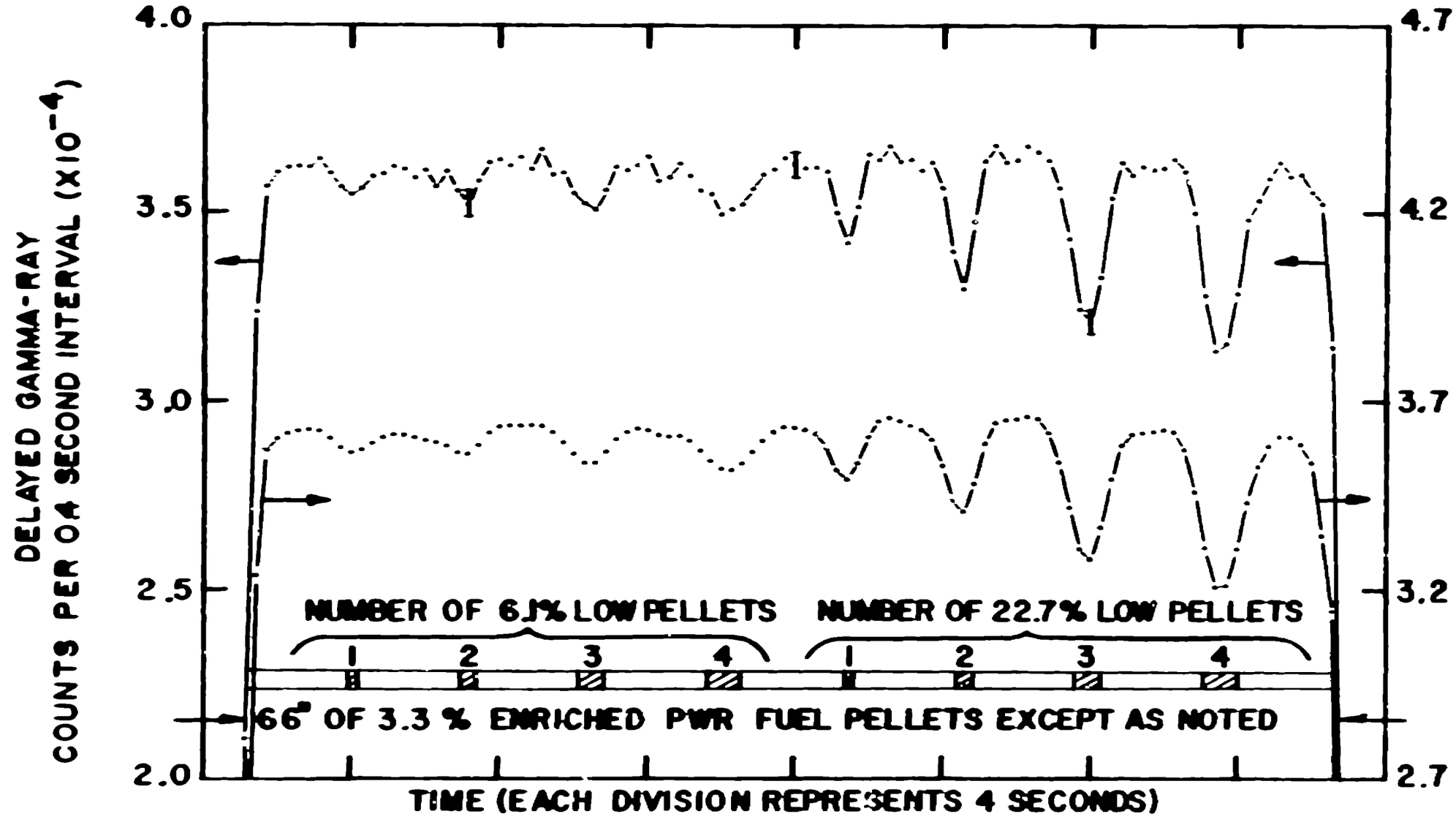

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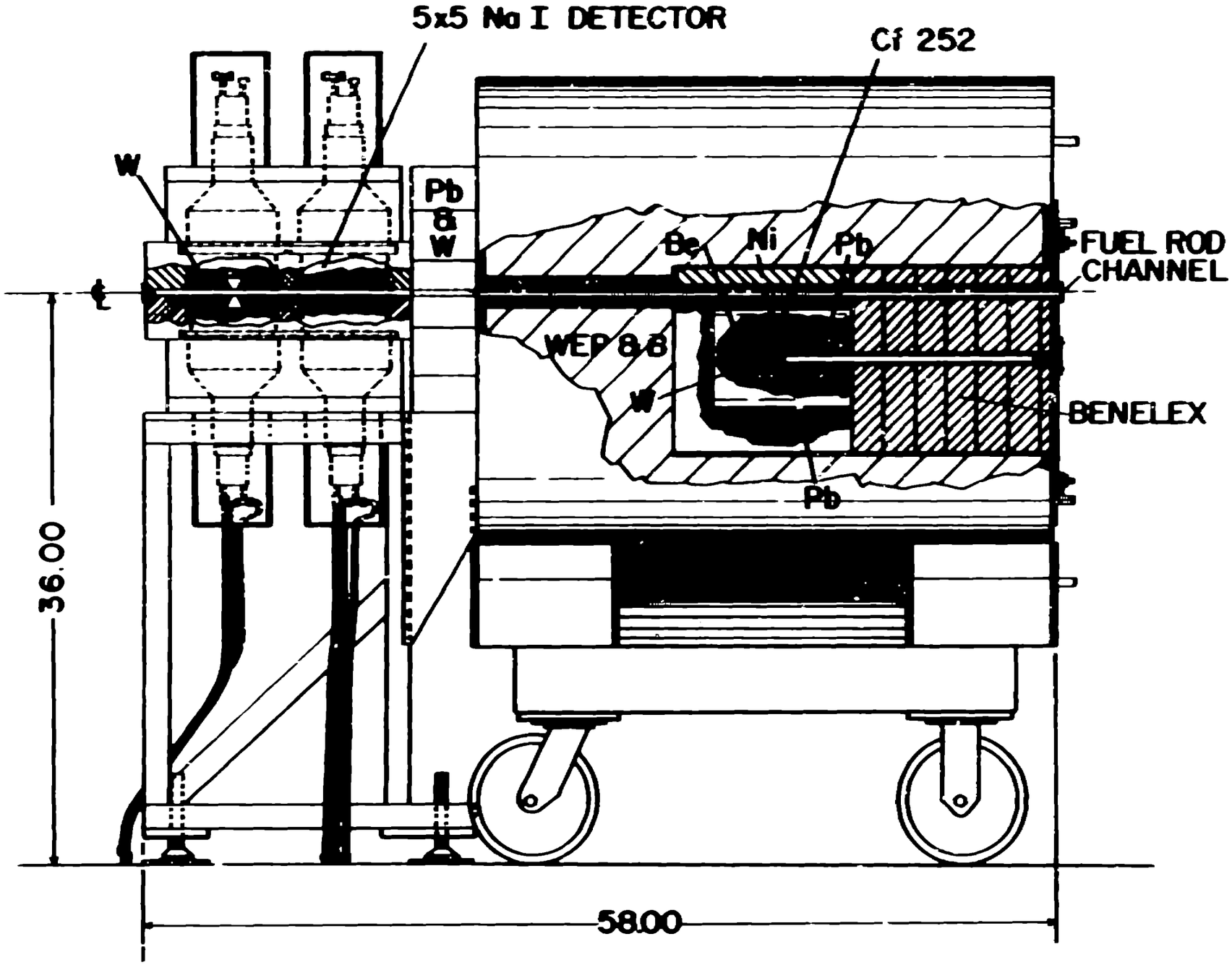

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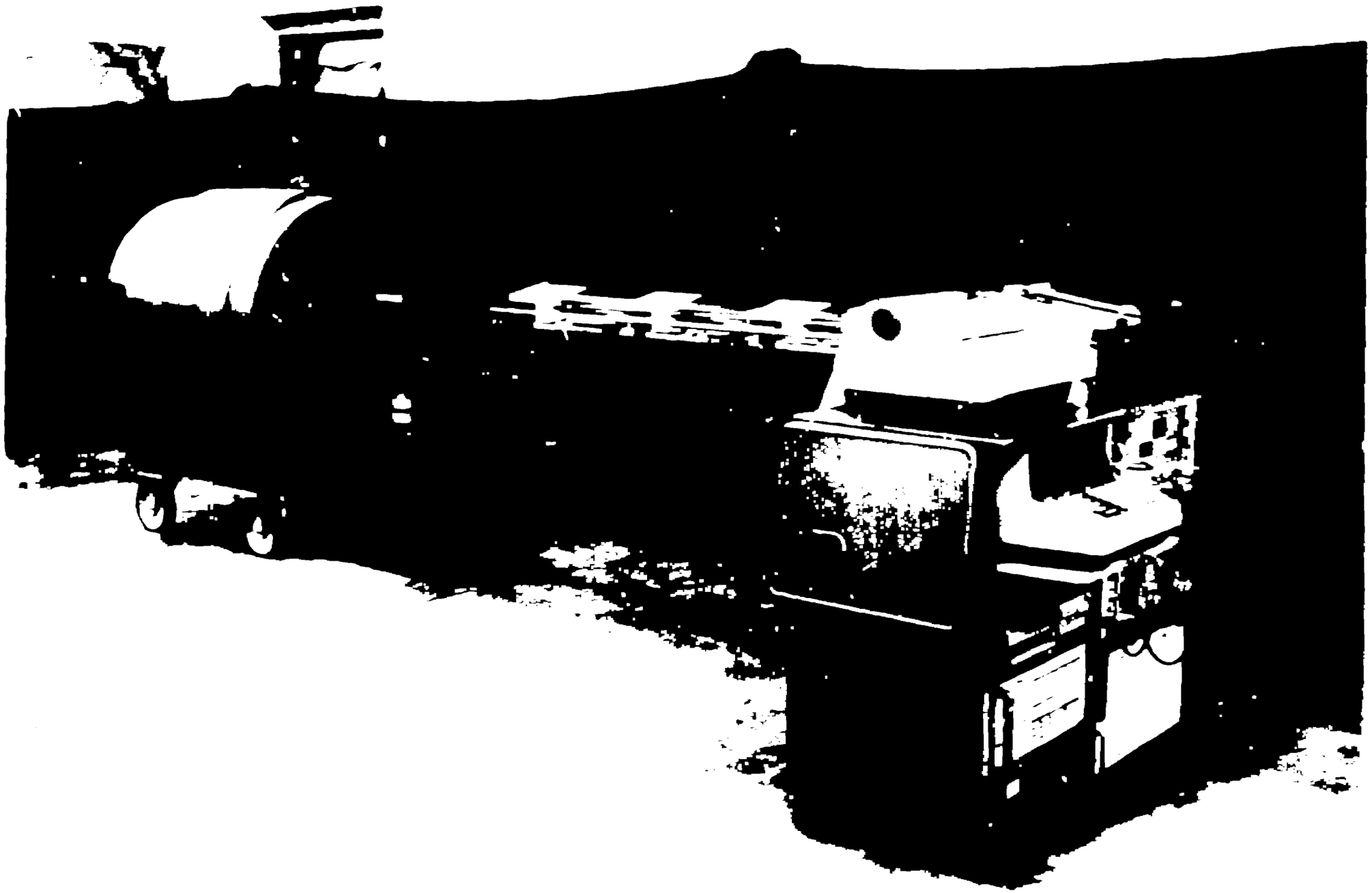

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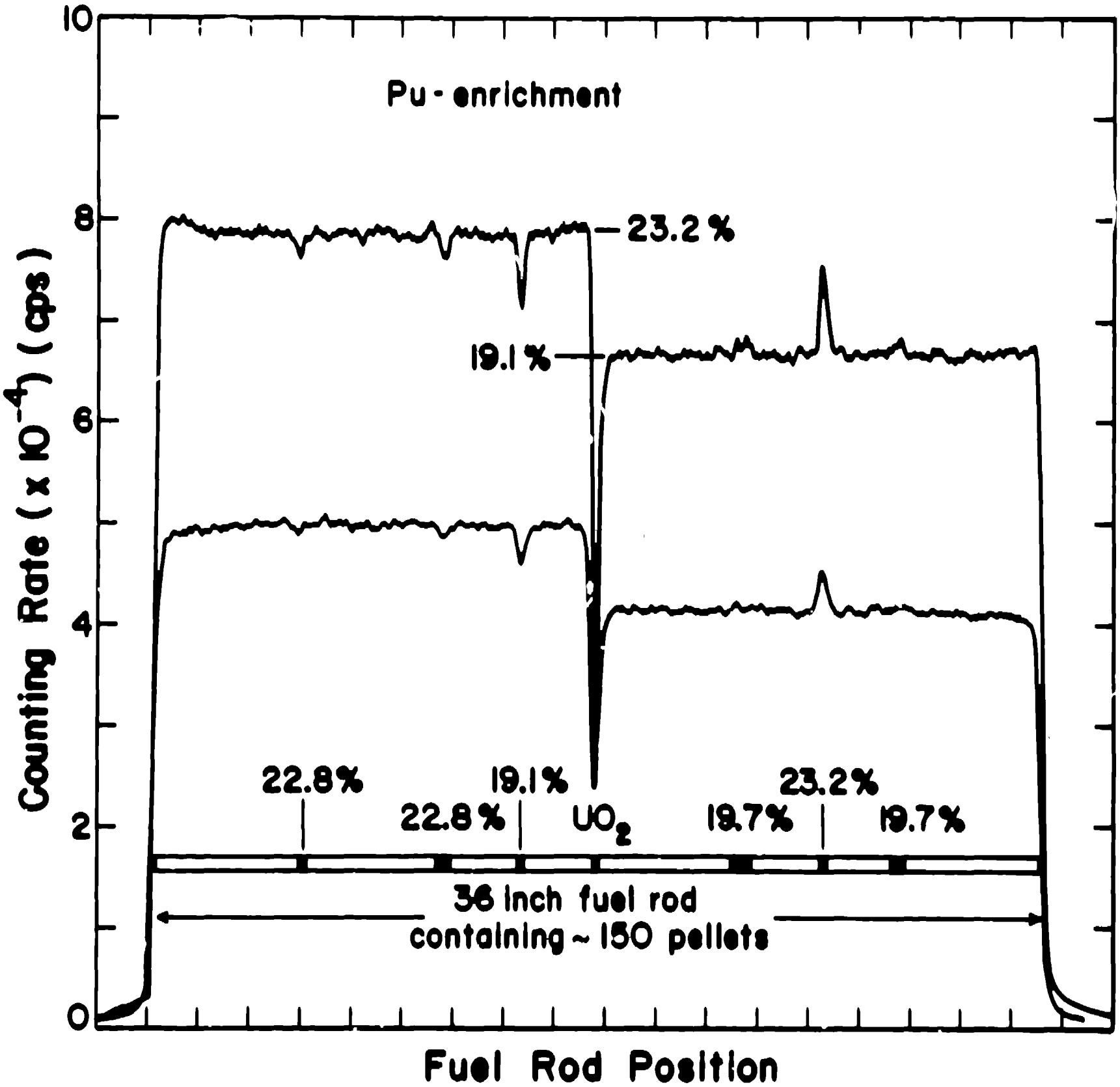

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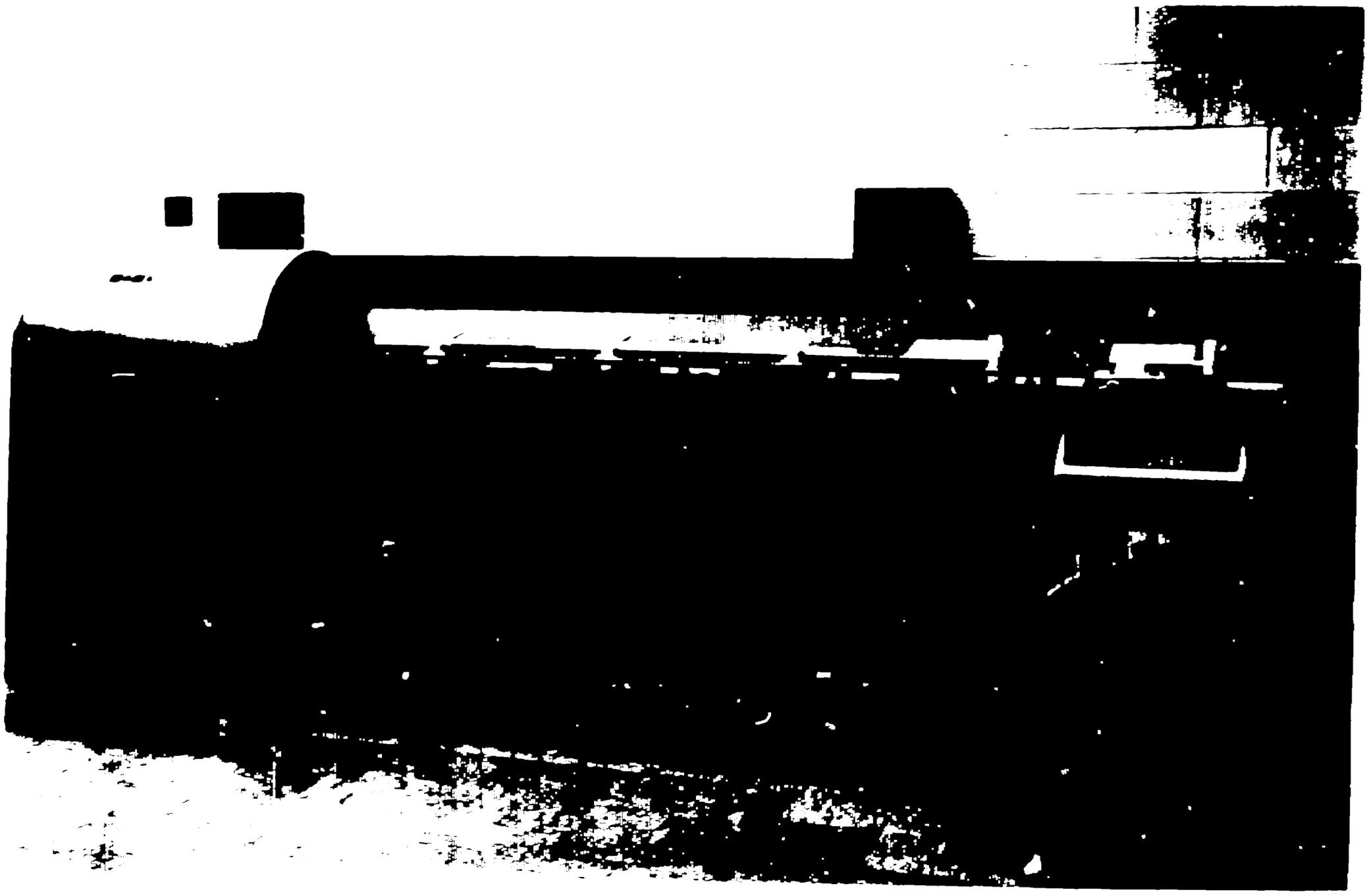

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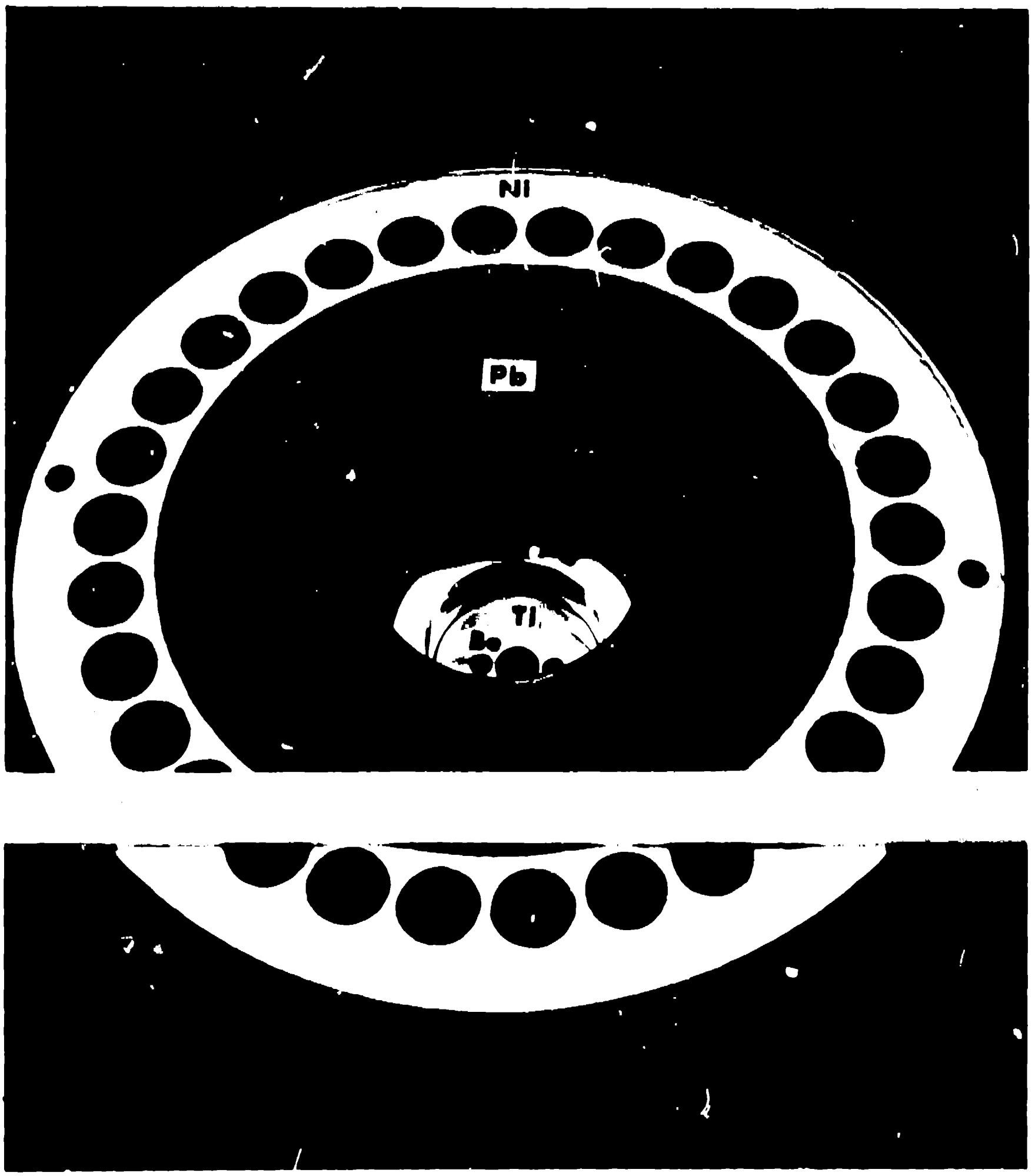

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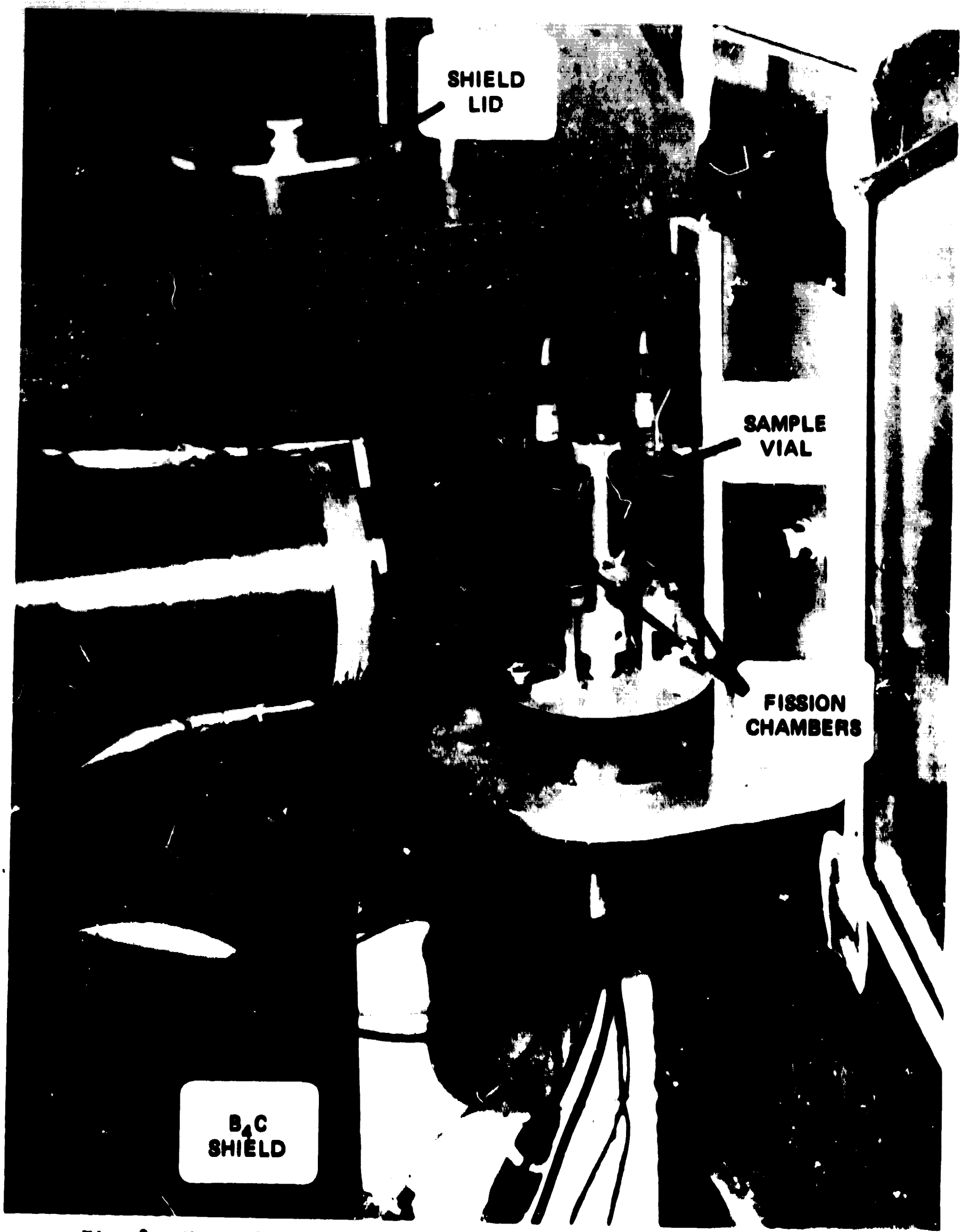
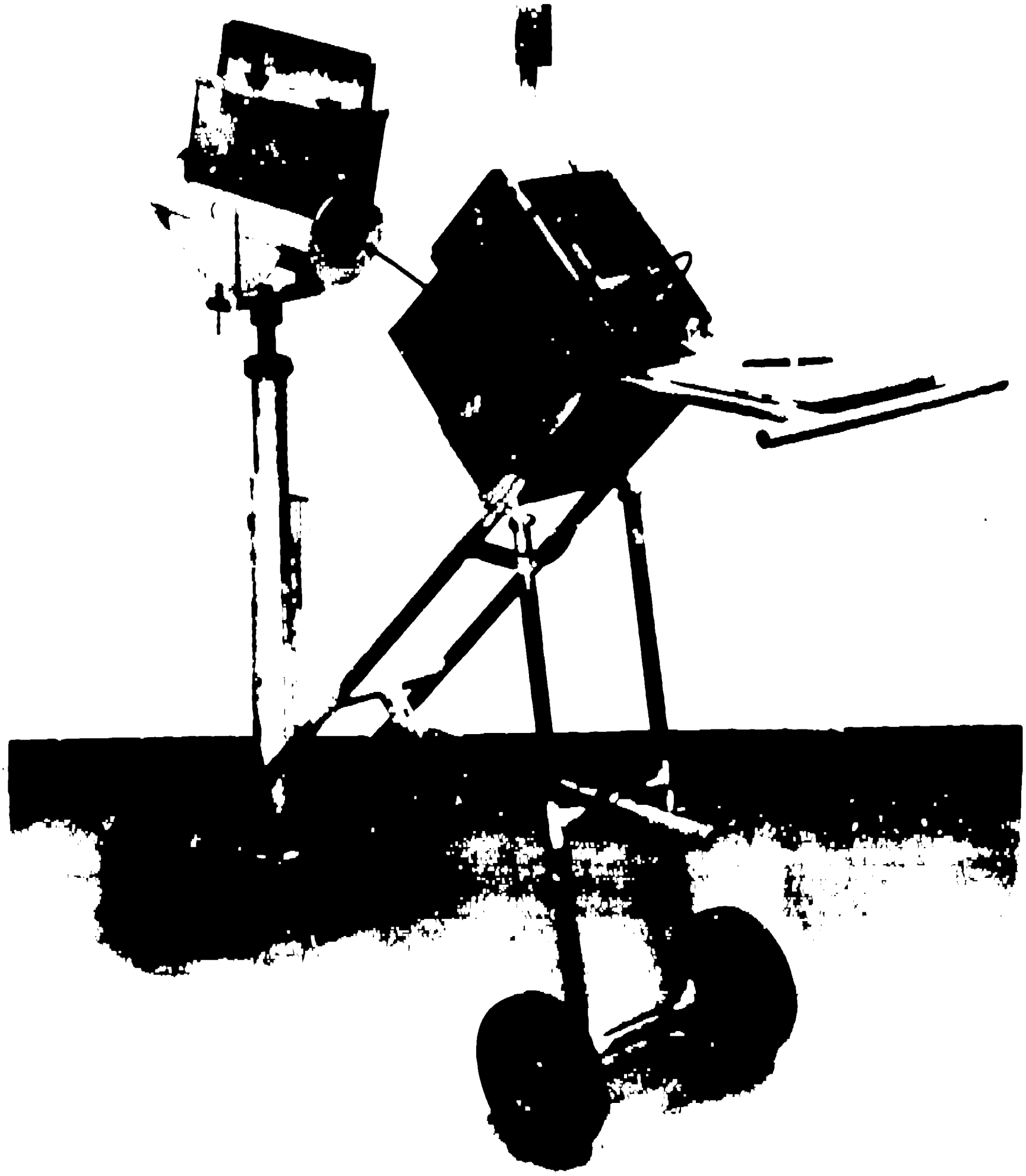


Fig. 9. View of small-sample assay station with B_4C shield removed.



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