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AUTHOR(S): J. W. Tape, M. P. Baker, R. Strittmatter, M. Jain, and M. L. Evans

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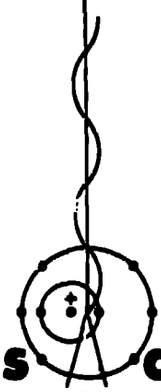

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SELECTED NONDESTRUCTIVE ASSAY INSTRUMENTATION FOR AN INTERNATIONAL SAFEGUARDS SYSTEM AT URANIUM ENRICHMENT PLANTS

J. W. Tape, M. P. Baker, R. Strittmatter,
M. Jain, and M. L. Evans
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

ABSTRACT

A selected set of nondestructive assay instruments for an international safeguards system at uranium enrichment plants is currently under development. These instruments are of three types: in-line enrichment meters for feed, product, and tails streams; area radiation monitors for direct detection of high-enriched uranium production, and an enrichment meter for spent alumina trap material. The current status of the development of each of these instruments is discussed, with supporting data, as well as the role each would play in a total international safeguards system.

I. INTRODUCTION

Uranium enrichment plants, like chemical separation facilities, are unique in the nuclear fuel cycle in that they change the strategic value of the nuclear materials that they process. The issues surrounding international safeguards for reprocessing plants have been extensively discussed during the past few years and, as is well known, there is not universal agreement among nuclear nations about the role of spent fuel reprocessing in the commercial fuel cycle. Enrichment plants, on the other hand, have not been in the news nearly so much (at least until this spring) and it is generally agreed that low-enriched uranium (LEU), produced by some kind of isotope separation facility, is necessary for a large-scale commercial nuclear power capability. In the past, most users of LEU have directly or indirectly purchased "enrichment services" from the few suppliers with isotope separation plants; however, in the near future it is expected that some nations may attempt to ensure their own supplies of enriched uranium by obtaining enrichment plants.[1] For these reasons enrichment plant safeguards is expected to become a greater concern of the international safeguards community.

Although a number of new techniques for separating uranium isotopes on a commercial scale are being developed throughout the world, in the next few years the bulk of the world's separative capacity will still be in gaseous diffusion plants. However, many new facilities will probably use gas centrifuge technology.[1,2] The increased use of centrifuges to enrich uranium increases the problems associated with safeguarding enrichment plants, and the safeguards strategies and

instruments discussed here are designed to be applied to a large-scale centrifuge facility.

An international safeguards system at an enrichment plant can use three primary strategies: material accounting on the declared feed, product, tails, and waste streams of the facility; containment and surveillance methods to ensure the validity of the accounting data obtained at the key measurement points; and methods that involve the direct detection of the production of high-enriched uranium (HEU). The first two strategies are applicable to detecting diversion of either LEU or HEU and the third is obviously intended to directly detect the use of some part or all of the separative capacity of the plant for the production of weapons-grade uranium. The application of these three strategies to safeguarding a centrifuge facility is both more difficult and at the same time more important than their application at a diffusion plant. Large separative capacity centrifuge plants are relatively easy to reconfigure for the production of HEU, and once reconfigured they can begin producing top product in less than one day.[3] Furthermore, the technology surrounding the centrifuge process is highly protected for reasons of national security, nonproliferation, and/or commercial advantage, and plant operators have been reluctant to allow international inspectors inside the cascade halls of their facilities. The application of effective international safeguards at a centrifuge plant is a difficult but, we feel, not impossible task; one that has potential technical solutions.

The instruments and methods described in the next section can be viewed in part as having application to all three of the safeguards strategies for enrichment plants; however, their primary functions are in the areas of material accounting and direct detection of HEU production. Containment and surveillance techniques such as personnel and vehicle monitors have been reported elsewhere.[4,5]

II. SAFEGUARDS MEASUREMENT PRINCIPLES AND PRELIMINARY RESULTS

The Los Alamos Scientific Laboratory (LASL) safeguards technology group (Q-1) is currently developing three different instruments as a part of the Department of Energy (DOE) program to develop a comprehensive safeguards system for enrichment plants; a gas phase enrichment meter, an alumina trap material enrichment meter, and a large array of neutron detectors to monitor the enrichment of a facility's gas phase inventory.

A. The Gas Phase Enrichment Meter

In-line enrichment meters in the feed and withdrawal (F/W) building of a centrifuge plant can provide an essentially continuous monitor of the enrichment of the UF_6 stream flowing into and out of the process. The enrichment data, when combined with on-line weights for the feed, product, and tails

cylinders, yield a real-time dynamic material balance for the plant. Although the accuracy of the in-line accountability data cannot compete with the accuracy of conventional off-line sampling and weighing data on a measurement-for-measurement basis, the 100% sampling and the timeliness of the in-line meter data make it valuable when applied to centrifuge plants. In-line monitors can be considered to be components of both material accounting as well as containment and surveillance systems.

An in-line enrichment meter for liquid UF_6 product has been developed and successfully operated at the Goodyear Atomic (GAT) diffusion plant since 1973.[6] This instrument, which measures a liquid stream at 3% enrichment, employs the gamma-ray enrichment principle and a NaI(Tl) gamma-ray detector and electronics system. It is possible to measure the enrichment of liquid UF_6 streams of any assay using this general technique if the NaI detector is replaced by a higher-resolution Ge detector.

Feed streams at enrichment plants are typically in the gas phase, and the enrichment principle employed in the GAT product monitor will not work unless a large thickness of gas is assayed (>1m); however, the gas phase enrichment can be determined by combining measurements of the ^{235}U concentration and the uranium concentration in the gas.

The ^{235}U concentration is related to the intensity of 186-keV gamma rays emitted by ^{235}U as measured by a gamma-ray detector that views a fixed volume of the UF_6 gas. The relationship between the enrichment, I, and the 186-keV gamma-ray count rate is given by the formula:

$$I = \frac{R(186)\ CF}{\rho\ k} \qquad (1)$$

where R(186) is the measured count rate, CF is the attenuation correction factor,[7] ρ is the density of UF_6, and k is a calibration constant. The attenuation correction, which accounts for the scattering and absorption of gamma rays in the gas, is a function of the elemental composition and density of the gas.

For UF_6 it is possible, in principle, to calculate both the attenuation correction and ρ from temperature and pressure measurements; however, the measurement of the attenuation of an external source of gamma rays transmitted through the gas provides a more independent means of determining these quantities and thereby determining I from Eq. (1).

J. C. Bailey at Oak Ridge[8] has measured the 186-keV counting rate as a function of the pressure of UF_6 gas in a

external ^{235}U source to determine the attenuation correction factors. We are developing an in-line gas-phase enrichment meter, shown schematically in Fig. 1, that employs an ^{241}Am transmission source to determine both the UF_6 gas density and the attenuation correction factor. The 60-keV ^{241}Am gamma-ray source has the advantage over the external ^{235}U source used by Bailey that it is easily resolved from the 186-keV gamma ray from ^{235}U and consequently it can be measured simultaneously with the 136-keV gamma ray.

Construction of a prototype instrument is not yet complete; however, we expect to be able to measure gas phase enrichments over a pressure range of approximately 200 to > 1000 Torr. The major problem associated with this method is likely to be background gamma rays from ^{238}U daughters and ^{235}U absorbed on the inside surface of the measurement chamber. The current design employs a NaI(Tl) gamma-ray detector and a computer-based multi-channel analyzer (MCA) to acquire the counting data as well as temperature and pressure data for the UF_6 gas. We are planning "proof of principle" experiments to determine the accuracy of the enrichment measurement as a function of gas density.

B. The Trap Material Enrichment Meter

The trap material enrichment meter is designed to indirectly detect HEU production by screening outgoing alumina trap material for enrichments greater than the declared top product of the facility. It can also possibly provide information on the ^{235}U content of the trap material for material balance purposes.

A number of assay techniques might provide the data required for the enrichment measurement of relatively low-density uranium-bearing waste. The thermal neutron coincidence counter method described below has been chosen because of its simplicity and the relative ease with which the instrument can be scaled in size to match the trap material containers in use at a particular facility.

Compounds of uranium and fluorine emit neutrons from two sources; spontaneous fission neutrons and (α,n) reactions in which an alpha particle from the decay of a uranium nucleus interacts with a ^{19}F nucleus and emits a neutron. The (α,n) neutrons are essentially all produced by the decay of ^{234}U and ^{238}U while spontaneous fission neutrons arise almost solely from the decay of ^{238}U nuclei.[9] (It should be noted that for recycled uranium in compounds with fluorine the ^{232}U and ^{236}U components present may contribute significantly to the neutron production rate.)

Since neutrons from fission are emitted simultaneously, coincidence counting techniques may be used to distinguish between neutrons produced from (α,n) reactions and those

resulting from spontaneous fission. The measurement of the ratio of the total neutron counting rate (N_T) to the coincident neutron counting rate (N_C) from a uranium-fluorine compound can then yield an approximate determination of the uranium enrichment provided the following assumptions are satisfied: 1) the ^{234}U (α,n) reactions dominate the total neutron production (N_T), 2) the ^{238}U spontaneous fission neutrons dominate the coincident neutron production (N_{SF}), and 3) the ^{234}U-to-^{235}U ratio does not vary widely.

Figure 2 shows the calculated total and spontaneous fission neutron rates and their ratio (N_T/N_{SF}) as a function of ^{235}U enrichment (I) assuming a constant $^{234}U/^{235}U$ ratio of 1/125. The values for N_T and N_{SF} were calculated for 5-kg masses. The ratio is not only a strongly increasing function of increasing enrichment, but is also independent of sample mass. Thus, an instrument that measures both total and coincident neutrons can give a measure of enrichment from the ratio of counting rates, and a measure of the ^{234}U mass (and indirectly the approximate ^{235}U mass) from the total neutron counting rate.

A thermal neutron coincidence (well) counter with a shift-register electronics package[10] can probably screen the enrichment of trap material samples as large as 55-gal drums using the principles discussed above. The "proof of principle" measurements were performed using a standard-size well counter (~10 ℓ) and small samples containing uranium-fluorine compounds. In the first of these measurements, eight 1S cylinders of UF_6 obtained from Goodyear Atomic Corporation (GAT) covering an enrichment range of 0.2-97.6% were analyzed. The measured ratio (N_T/N_C) for these samples is shown in Fig. 3 as a function of enrichment for various well-counter liner materials. The measured results behave as expected for enrichments up to about 20% where the ratio reaches a maximum and then actually decreases.

The deviation of the ratio behavior above 20% enrichment is due primarily to (α,n) neutron-induced fission of the ^{235}U in the samples. In the simple picture of Fig. 2, coincident neutrons arise only from ^{238}U spontaneous fissions and the expected coincidence neutron counting rate per gram of uranium decreases as the ^{235}U content increases. The addition of coincident neutrons from induced fissions actually results in an increase in the coincidence counting rate as a function of increasing ^{235}U enrichment for these UF cylinders. A semiempirical formula, in which the induced fission component of the coincidence counting rate is taken as proportional to the product of ^{234}U and ^{235}U masses in the samples, fits the observed data adequately.

Measurements of alumina trap material samples, in which the uranium is widely dispersed, are not expected to exhibit this

(α,n) multiplication effect to as large a degree as the measurements of the more compact UF_6 cylinders described above. Nevertheless, it is valuable to use the UF_6 samples as a "worst case" situation and attempt to minimize the multiplication effects. Such attempts necessarily must focus on the materials that line the cavity of the well counter since induced fissions that occur prior to the escape of the initiating neutron from the sample can only be affected by changes in sample geometry or composition. Figure 3 shows the measured results for N_T/N_C for no liner, a Cd liner, and a liner made up of Cd and boral. Although the changes are not dramatic, discrimination between typical product enrichments (~3%) and higher enrichment values appears to be improved for the Cd-boral liner.

A second series of well-counter measurements (using no special liners) was made on six samples of alumina trap material obtained from Union Carbide and Goodyear Atomic. The uranium contained in these samples had three enrichments in the range ~.2%-17% and weight concentrations of 1% and 4%. Each sample (14-15 kg of alumina), which was the content of one full trap, was split into two or three smaller samples that were compatible with the sample cavity of the well counter. The measured ratio (N_T/N_C) is shown in Fig. 4 and again shows a sizable increase with increasing enrichment. The counting rate ratio at a given enrichment was independent of the uranium loading with the possible exception of the 17% enrichment traps where a small multiplication effect may have been observed. Thus, for these sample sizes and uranium loadings, this technique can clearly be used to "flag" high-enrichment material.

C. Neutron Emission Monitoring Technique (NEMTech)

One of the primary goals of an enrichment plant safeguards system should be the ability to directly detect the production of HEU by that facility. This is especially true for centrifuge plants where process equilibrium times can allow for the production of significant quantities of HEU in a short period of time. Material accounting methods and containment and surveillance techniques provide major deterrents to HEU production by monitoring the flow of uranium into and out of the plant; however, if these measures are defeated by some means the operator can divert all or part of the separative capacity of his plant to HEU production with little chance of being detected while the operation is in progress. Instruments such as the trap material enrichment meter can provide valuable evidence of HEU production, but the information may not be timely enough for international response. Detectors that could directly monitor the operation of enrichment cascades, even under conditions of limited inspector access or no access to the cascades, would be extremely valuable additions to the total safeguards system.

Area radiation monitors have the potential to detect production of HEU, as both the gamma-ray and neutron-emission rates from UF_6 increase with increasing enrichment (see Fig. 2). The poor penetrability of the primary gamma rays from uranium tends to rule out the use of gamma detectors for area radiation monitors. Neutron detection schemes have a dual advantage in that the radiation penetrates matter relatively easily (and thus is difficult to shield) and the monitors can be made up of large arrays of simple detectors.

Field measurements of neutron-emission rates in and around gaseous diffusion plants reported by Walton[11] show that the neutron intensity inside a diffusion cascade does reflect the operation of that cascade. Monitoring centrifuge cascades is much more difficult because the smaller gas phase inventory of the centrifuge process as compared to the diffusion process results in a smaller signal and poorer signal-to-background ratio. In principle, large arrays of polyethylene-moderated ^3He neutron detectors could be deployed either inside the cascade halls (under conditions of limited inspector access to those areas) or on the facility roof. Each detector in the array monitors the neutron emission rate, and therefore the average enrichment of the UF_6, over a limited region of the cascade area. If the average enrichment over the area viewed by a detector (or group of detectors) increases, the detector(s) count rate will increase as compared to other detectors in the array that are viewing normal operation.

Although NEMTech is very attractive on paper, many aspects of the method need to be proved prior to its application at a centrifuge plant. We are currently undertaking computer model studies of centrifuge facilities with detector arrays placed on the roof and inside the cascade to try to determine if the arrays have the basic sensitivity to detect various HEU production schemes. The results of a very simple model are shown in Fig. 5. In this model detectors #1 and #2 each have an active area of $1 m^2$ and an average efficiency for detecting neutrons of 10%. Each detector is placed on the roof of a separate $2100-m^2$ cascade building with a gas phase inventory of 40 kg. The detectors are isolated from each other so that there is no cross talk. The background rate in each detector is taken to be equal and constant at 500 counts per minute (cpm). Detector #1 always views a gas volume with an average enrichment of 0.7% while the enrichment viewed by Detector #2 varies from 0.7% to 15%. Fig. 5 shows the difference in the total counts accumulated by the two detectors, for 20-min and 1-h count times, as a function of the enrichment viewed by Detector #2. The "measurements" are dominated by the background; however, even in a 20-min count time the total counts accumulated by Detector #2 viewing 10% enrichment [illegible] those detected by #1 by more than two standard deviations. The assumed background rate is based on rates measured by unshielded detectors around an operating enrichment [illegible]

can possibly be improved by the use of shielding. On the other hand, background rates may fluctuate from one detector location to another and thereby complicate this monitoring scheme. If the results of the computer simulation studies continue to be encouraging we will build an experimental array of detectors for field test and evaluation.

III. CONCLUSION

The three instrument systems described here represent only a part of the efforts at LASL and elsewhere to develop the components of an effective international safeguards system for large-scale centrifuge enrichment plants. Many questions remain to be answered in this program, ranging from the technical feasibility of some of the proposed methods, such as NEMTech, to the effectiveness of any given integrated safeguards system in detecting or deterring the diversion of either LEU or HEU at a centrifuge facility. We anticipate that some of the unanswered technical questions relating to the three methods dicussed in this paper will be resolved during the next year's research and development activities and that these instruments will then be extensively tested in the field under realistic operating conditions.

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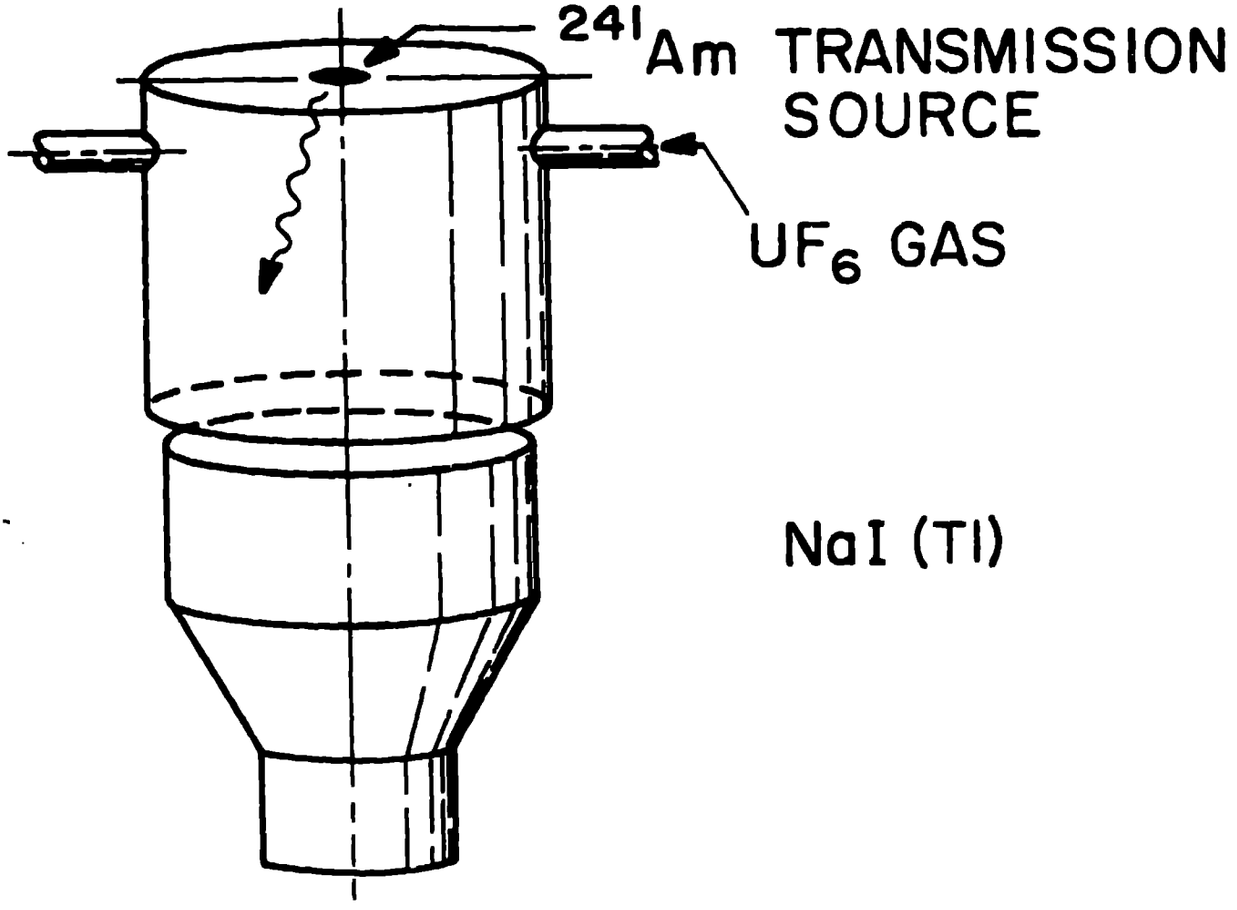
FIGURE CAPTIONS

1. Conceptual design of a gas-phase enrichment meter. The intensity of gamma rays emitted by $^{235}UF_6$ and the attenuation of gamma rays transmitted through UF_6 gas are measured and the resulting data are used to calculate the ^{235}U enrichment, as described in the text.
2. Neutron emission rates for a 5-kg mass of UF_6 vs ^{235}U enrichment. The dot-dashed curve labeled N_T is the total number of neutrons emitted per second. The dashed curve labeled N_{SF} is the number of spontaneous fission neutrons emitted per second. The solid curve is the (mass-independent) ratio of total neutrons emitted to spontaneous fission neutrons emitted. A constant $^{234}U/^{235}U$ ratio of 1/125 has been assumed.
3. Thermal neutron coincidence counter data for 15 cylinders of UF_6. The ratio of total neutron counts to coincidence counts (N_T/N_C) is plotted vs the ^{235}U enrichment of the samples for various coincidence counter liner materials.
4. Thermal neutron coincidence counter data for alumina trap material standards loaded with 1% and 4% UF_6 by weight. The ratio N_T/N_C is plotted as a function of the ^{235}U enrichment of the contained uranium.
5. Δ vs the average enrichment viewed by Detector #2. Δ is the difference in accumulated counts observed in two detectors placed on the roof-tops of two separate gas centrifuge enrichment cascades. The average enrichment of the gas phase inventory viewed by Detector #1 is 0.7% while the enrichment of the gas viewed by Detector #2 varies from 1.7% to 15%.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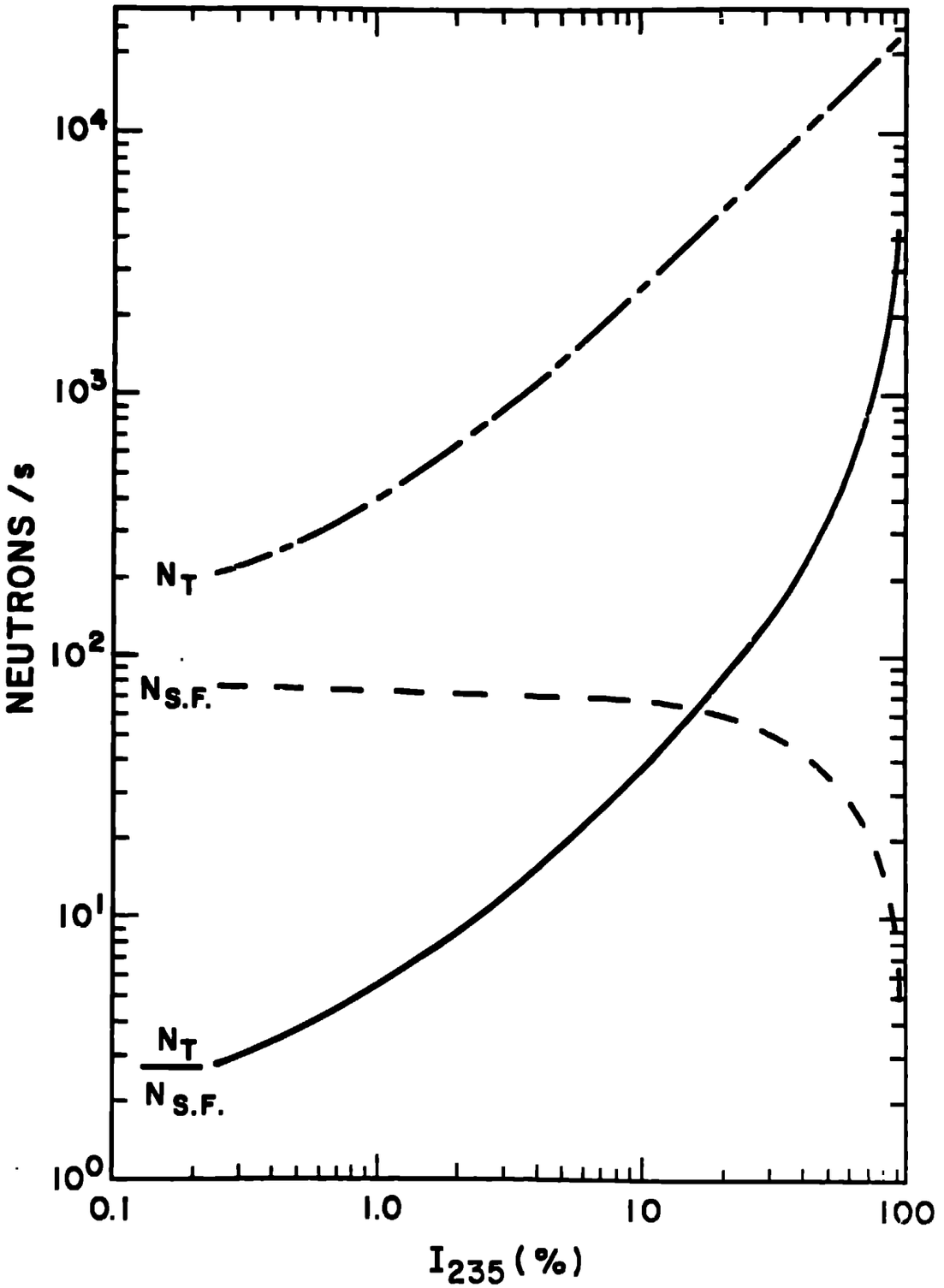
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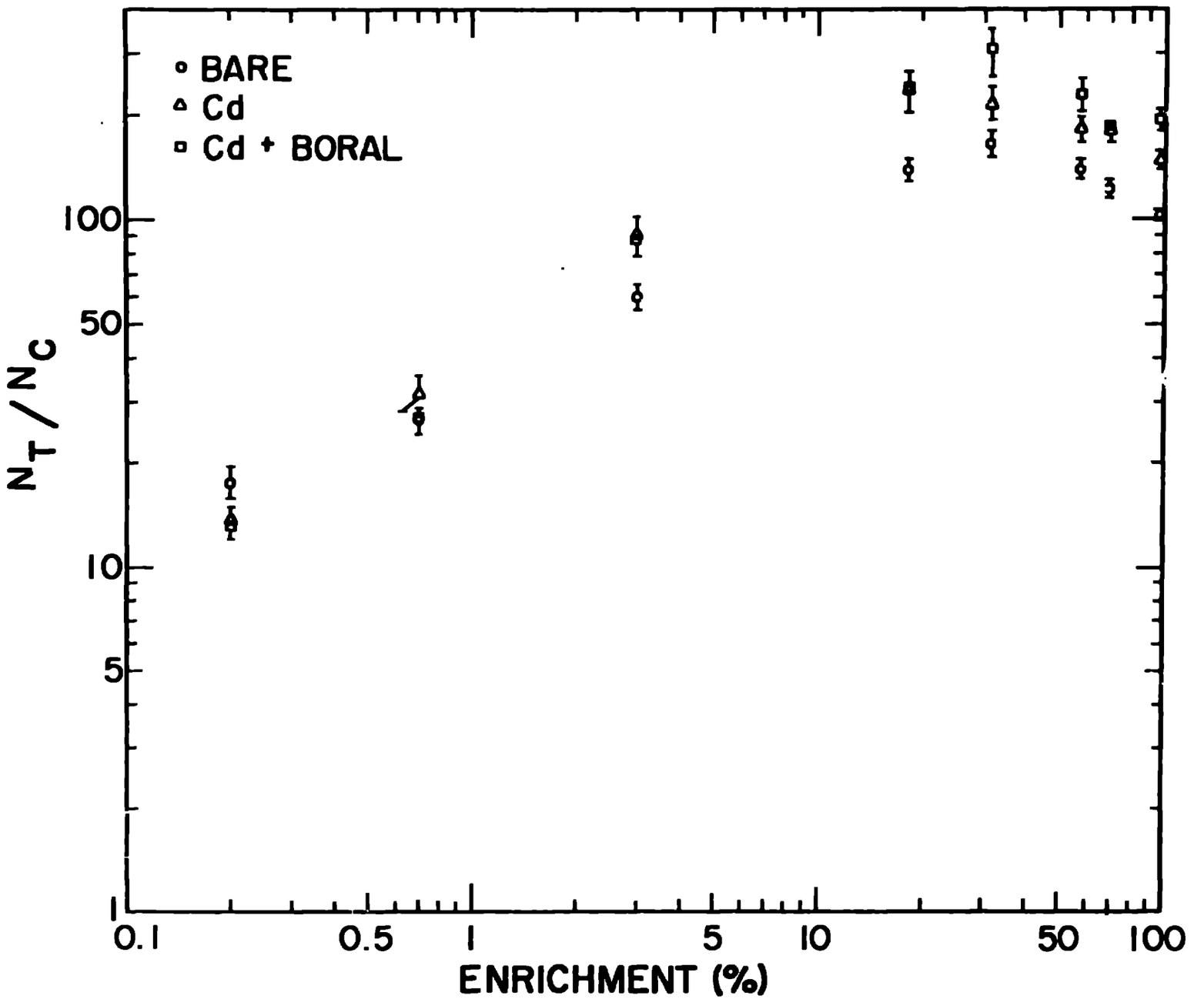
I_{235} (%)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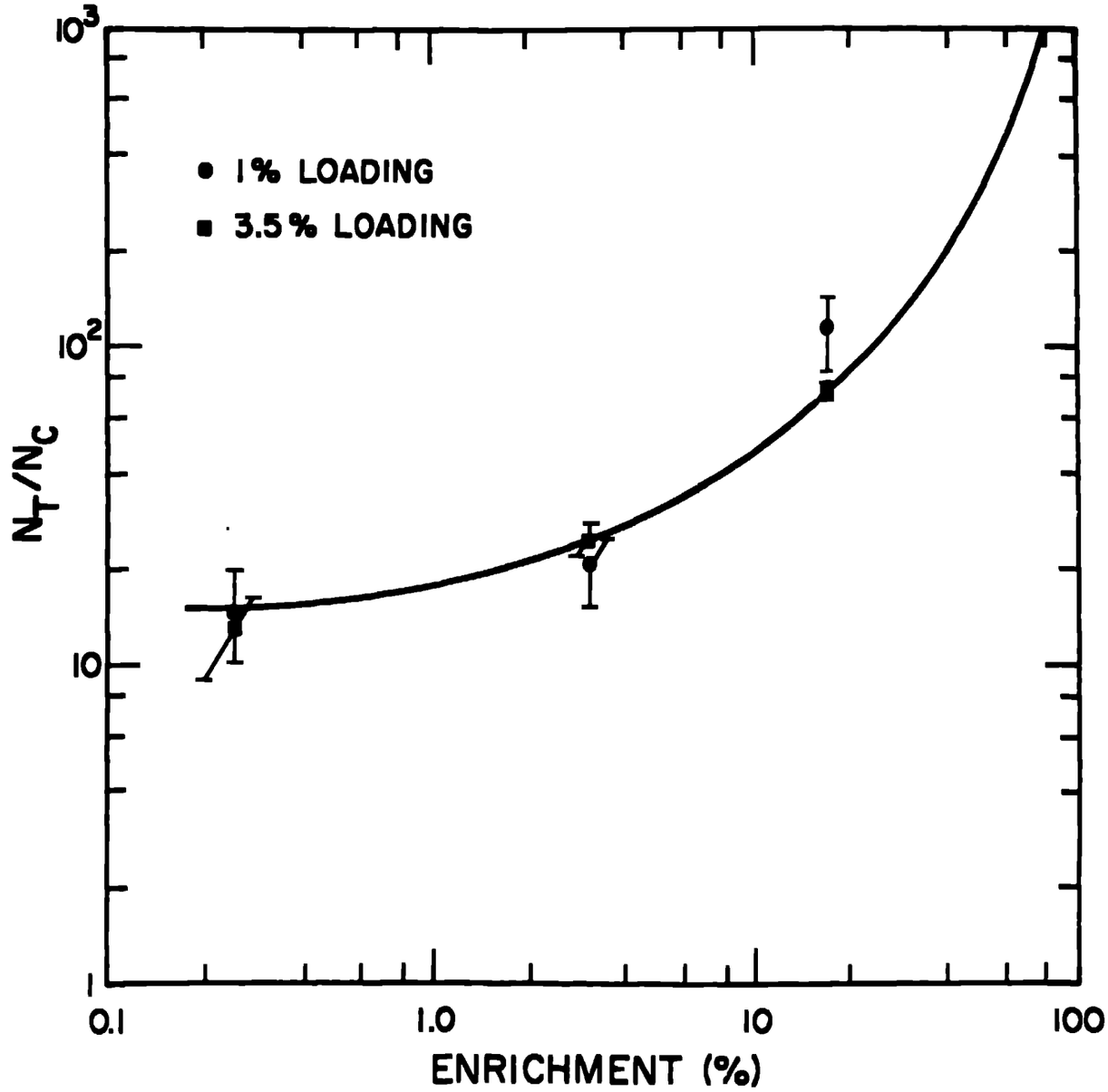
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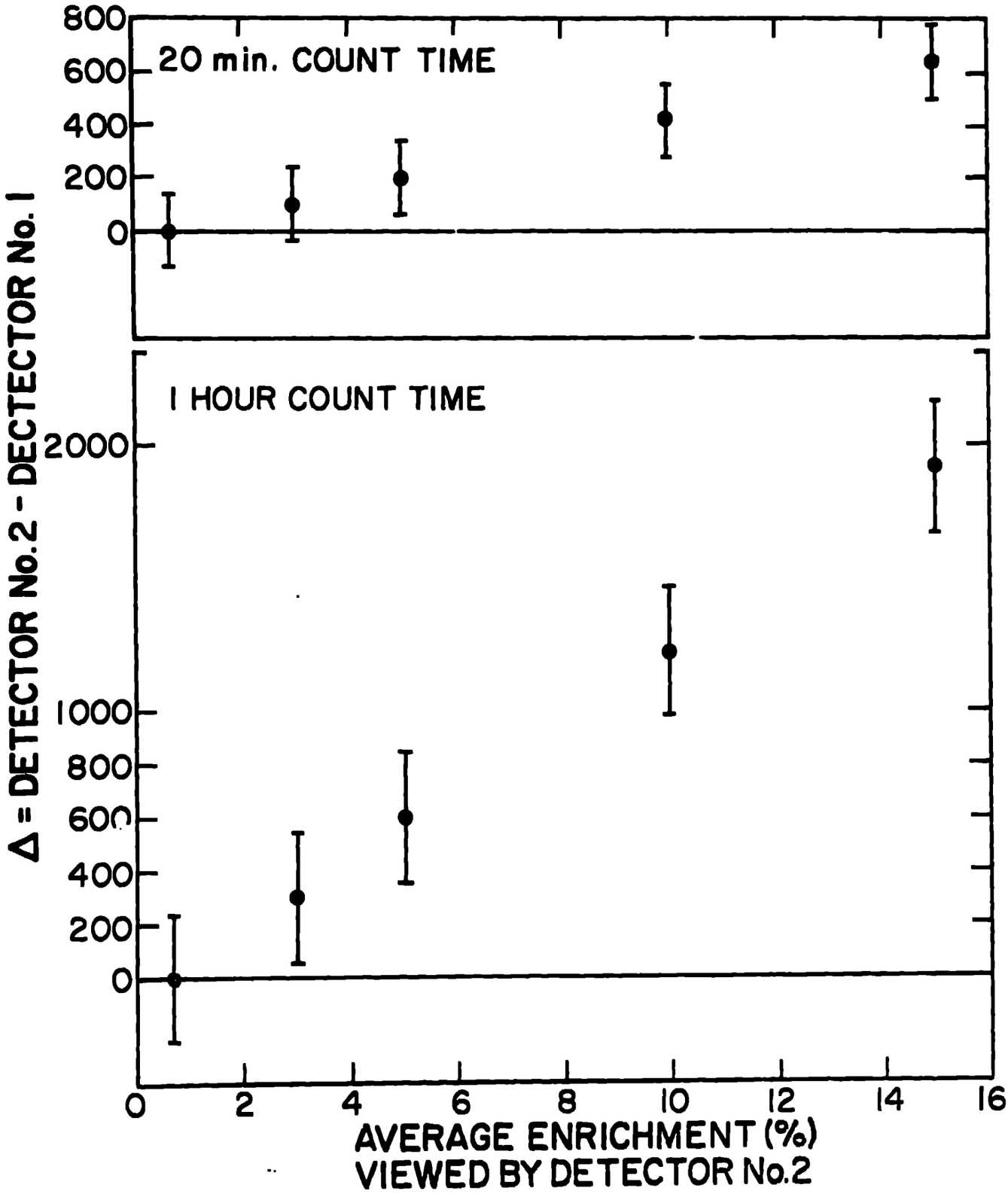
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1 HOUR COUNT TIME
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16
AVERAGE ENRICHMENT (%)
VIEWED BY DETECTOR No.2