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# ADVANCES IN PASSIVE NEUTRON INSTRUMENTS FOR SAFEGUARDS USE

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# ADVANCES IN PASSIVE NEUTRON INSTRUMENTS FOR SAFEGUARDS USE*

## ABSTRACT

Passive neutron and other nondestructive assay techniques have been used extensively by the International Atomic Energy Agency to verify plutonium metal, powder, mixed oxide, pellets, rods, assemblies, scrap, and liquids. Normally, the coincidence counting rate is used to measure the $^{240}$Pu-effective mass and gamma-ray spectrometry or mass spectrometry is used to verify the plutonium isotopic ratios. During the past few years, the passive neutron detectors have been installed in plants and operated in the unattended/continuous mode. These radiation data with time continuity have made it possible to use the totals counting rate to monitor the movement of nuclear material. Monte Carlo computer codes have been used to optimize the detector designs for specific applications. The inventory sample counter (INVS-III) has been designed to have a higher efficiency (43%) and a larger uniform counting volume than the original INVS. Data analyses techniques have been developed, including the "known alpha" and "known multiplication" methods that depend on the sample. For scrap and other impure or poorly characterized samples, we have developed multiplicity counting, initially implemented in the plutonium scrap multiplicity counter. For large waste containers such as 200-L drums, we have developed the add-a-source technique to give accurate corrections for the waste-matrix materials. This paper summarizes recent developments in the design and application of passive neutron assay systems.

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## 1. INTRODUCTION

Neutron coincidence counting has been used extensively during the past two decades for the nondestructive assay (NDA) of nuclear material. The technique is useful primarily because fast neutrons penetrate well, and time-correlated neutrons are related to the fission process and thus the nuclear material content.

The most important advance during the past few years in the application of NDA for inspection verification was the use of passive neutron systems in the unattended/continuous mode of operation. The passive neutron systems have been used extensively in mixed oxide (MOX) fabrication facilities with excellent results for both reliability and accuracy. Inspection days in the field have been significantly reduced by the successful application of unattended NDA systems.

Computer calculations employing Monte Carlo codes have been used to optimize the design of $^3$He neutron-coincidence detector systems. Neutron coincidence counters have been applied to the assay of a wide range of plutonium masses and container sizes, making it necessary to emphasize different parameters to achieve specified detector characteristics.

Complicating the measurements using the simple spontaneous fission coincidence approach are additional coincidence neutrons from *induced fission* reactions in the plutonium. These induced fissions can be triggered by neutrons that are random in time [$(\alpha,n)$ neutrons] or by neutrons that originated from previous fission events (spontaneous or induced).

Equations that describe these multiplication effects have been developed by K. Bohnel,[1] N. Ensslin,[2] and others. In this paper, we describe methods that have been developed for assaying samples ranging from pure plutonium to impure MOX including scrap and waste.

## 2. CONTINUOUS/UNATTENDED MODE NDA

Large, automated facilities for fabricating plutonium fuel present both difficulties and opportunities for improved control and inspection of nuclear materials. The traditional methods of sample measurements requiring the transfer of the sample from the production line to the measurement station are not possible in most cases. The robotics used for automation require special containers for nuclear material that cannot be easily removed from the production line. Also, safety and radiation protection considerations require that the measuring devices be installed in the fuel production lines because, in general, personnel cannot be in the fuel-handling area with nuclear material. These operational constraints are common in many of the modern facilities that have been designed for fabricating and processing plutonium fuel.

To accommodate these facility features and to reduce the inspector's workload, we have designed the NDA equipment to be automated, amendable to unattended operation, and with a size and fuel mass capability to match the robotics fuel manipulators. Authentication techniques have been incorporated into the NDA systems so that the data can be used by independent inspectors.

The standardized containers and programmed fuel movements in automated facilities make it possible to perform more accurate NDA measurements than are possible in conventional nonautomated facilities. The NDA instrumentation can be custom designed and optimized for the particular measurement goal in the automated facility.

At the Plutonium Fuel Production Facility (PFPF) in Japan, passive neutron instruments have been installed to give complete measurement coverage of all the plutonium in the facility.[3] Figure 1 shows the instruments and the measurement locations within the plant. In addition to the input and output locations, more difficult to access locations such as glove boxes, process equipment, and waste containers are included in the measurement coverage.

The material categories that are measured include the input MOX powder, the output fuel assemblies, and waste drums. The process-line MOX powder, pellet trays, and scrap are measured inside the glove-box lines using detectors outside the glove boxes. The MOX holdup in glove boxes, furnaces, and process equipment is measured using large slab detectors on the outside of the equipment. Small grab samples destined for chemical analysis also are measured in the NDA systems before analyses.

After multiplication corrections, the coincidence neutron yield is directly proportional to the $^{240}$Pu-effective mass. Thus, by measuring the coincidence neutron yield from the plutonium in the facility, we can verify the entire plutonium inventory. Of special significance for PFPF is the capability to make routine quantitative measurements of the holdup and waste materials.

The passive neutron systems have been reliable with no failure leading to loss of inspection data during the initial three years of use. The accuracy and precision of the systems that are installed in the automated facility are better than can be obtained with portable NDA equipment. Repeat measurements with the $^{252}$Cf control sources have demonstrated a precision of 0.1% (standard deviation) without any normalization over the initial two years of operation.

Passive neutron measurement systems that operate in the unattended/continuous mode have been developed for the Siemens MOX-II facility in Germany. This instrumentation is designed for joint use by the IAEA and the EURATOM-Luxembourg inspectorate. These measurement systems were designed for the pellet storage area and the fuel rod assembly area. They combine neutron coincidence counting with electro-mechanical sensors to enhance verification assurance. The neutron system has a sensitivity of less than one pellet on a tray.[4] The Siemens NDA systems are scheduled for installation in 1994.

## 3. NEUTRON COINCIDENCE ANALYSES

For simple coincidence counting, there are two observables—the totals rate $T$ and the coincidence rate $R$. However, there are three unknowns—the effective spontaneous-fission mass $M_{240}$, the ($\alpha$, n) rate, and the neutron multiplication $M$. Thus, to solve for the plutonium mass, we must assume one of the variables is known [such as $\alpha$, where $\alpha$ is the ($\alpha$, n) to spontaneous-fission neutron ratio]. For those samples that are not pure, we get an error because the actual $\alpha$ is larger than the assumed $\alpha$.

The multiplication-corrected results using this known $\alpha$ approach[2] are more precise than any of the other neutron methods when the basic chemical-purity assumption is valid. This is the approach normally used with the high-level neutron coincidence counter family of instruments.

In the case of impure plutonium, the known $M$ approach[5] usually will give a more accurate answer than the known $\alpha$ approach. The question is how closely can we estimate $M$? The value of $M$ is a function of moisture, density, size, and shape; density and moisture are the most difficult parameters to determine. For the special case of fuel assemblies, pellets, and fuel pins, the moisture, size, shape, and density are known and $M$ can be accurately predicted. For cans of powder, we can always assume a nominal density that is similar to the calibration standards and obtain $M$ from the calibration curve of $M$ vs $^{239}$Pu-effective.[5]

This method using the known $M$ to solve for the mass is very attractive for samples with low plutonium density because $M$ approaches unity. For example, a can containing 0-100 g of plutonium in a 1-L volume will have $M$ range from only 1.00 ~ 1.01 and the uncertainty in $M$ is small.

The known $\alpha$ approach is currently applied in the IAEA's software for the HLNC. The same basic equations and data can be used for the known $M$ approach;[6] however, modified software is required to obtain the plutonium mass from the known $M$ calibration. The current software defaults to $M = 1$ for cases where the $R$ and $T$ data result in a value of $M \leq 1$. This is essentially a special case of the known $M$ approach for $M = 1$.

If the results from both the known $\alpha$ and known $M$ analyses were routinely made available to the inspector, outlier samples could be easily identified and more accurate assay values could be obtained by supplemental knowledge related to the sample's purity and physical characteristics.

For cases with unknown multiplication and unknown purity, we have made use of the higher neutron moments with multiplicity counting[7,8] to obtain more accurate assays.

## 4. MULTIPLICITY COUNTING

For bulk samples of plutonium that are impure, we need an additional observable to solve for the induced fissions and the unknown $\alpha$. The multiplicity counting gives us singles, doubles, and triples and thus allows the assay of samples with unknown purity and unknown multiplication. The theory and equations for multiplicity counting can be found elsewhere.[7,8,9]

For high-precision measurements of the triples rate, the neutron detector must have high efficiency and a reasonably short neutron die-away time. We have developed several multiplicity detectors[10] with efficiencies in the range of 50-60% including the plutonium scrap multiplicity counter[11] (PSMC) that was deployed for field use by the IAEA in 1992.

The PSMC evolved from multiplicity neutron detectors, developed at Los Alamos for impure plutonium samples. The new unit was designed to be more compact and to use fewer $^3$He tubes to obtain a high efficiency. We designed the PSMC by using the Monte Carlo Code for Neutron and Photon Transport (MCNP) to perform the Monte Carlo neutron calculations. The design goals for the PSMC were to obtain high efficiency (primary importance), uniform efficiency vs sample height, small die-away time, flat energy response, minimum number of $^3$He tubes, and minimum overall size and weight.

The MCNP calculations provided data to significantly improve on the previous designs. For example, the pyrochemical detector[10] used 126 helium 3 tubes to obtain an efficiency of 57%, whereas the present design uses 80 helium 3 tubes to get an efficiency of 55%, but with slight degradations in the die-away time and the energy response.

Figure 2 shows a schematic diagram of a PSMC with the $^3$He tubes surrounding the sample cavity with a diameter of 20 cm. The outer dimensions of the polyethylene ($CH_2$) shield are 66 by 66 by 80 cm. The total height is 92 cm. With the exterior polyethylene shielding, the weight of the unit is ~330 kg so the unit is not portable and it is normally left in the facility.

The initial results using the PSMC have been good and many scrap samples have been removed from the outlier category.

## 5. INVENTORY SAMPLE COINCIDENCE COUNTER

For small inventory samples containing only a few grams of $PuO_2$ or MOX, the *uncertainties* in the multiplication are negligible and accurate assays can be made for both pure and impure materials. Prior to the development of the INVS-III,[12] the primary uncertainties in measuring the small samples originated from counting statistics and sample positioning. These errors have been greatly reduced by the INVS-III with an efficiency of 43% and a uniform counting zone 10 cm long. With careful sample control, the INVS-III can give accuracies competing with destructive analysis.

The INVS-IV[13] is similar to INVS-III except the bottom end plug is replaced with a high-purity germanium detector for the simultaneous measurement of the neutron yield and the plutonium isotopic ratios.

Monte Carlo calculations were used to design the INVS-III to achieve the high efficiency and flat axial response. The detector has 18 helium 3 tubes with active lengths of 39.4 cm and a gas pressure of 6 atm. The end plugs contain graphite and the outside diameter is 30.8 cm. The neutron pulses are collected and pre-processed in three banks of AMPTEK amplifiers.

For some applications, the INVS-III has been positioned around an access pipe at the bottom of a glove box so that it is not necessary to remove the sample from the glove box for verification.

Recent work by EURATOM-Luxembourg[13] has demonstrated the ability of the INVS-III to measure small plutonium powder samples with a standard deviation of less than 0.2%. On-site laboratories are being designed with high-accuracy NDA systems to reduce the necessity of shipping plutonium inventory samples to chemical analysis laboratories.

## 6. WASTE DRUM ASSAY WITH THE ADD-A-SOURCE FEATURE

We have developed a new, passive-neutron measurement technique to improve the accuracy and sensitivity of the NDA of plutonium scrap and waste. The 200-L-drum assay system uses the classical NDA method of counting passive neutron coincidences from plutonium but has added the new features of "add-a-source" to improve the accuracy of matrix corrections and statistical techniques to improve the low-level detectability limits. The add-a-source technique[14] introduces a small source of $^{252}$Cf (~3 x $10^4$ n/s) near the external surface of the sample drum; the drum's perturbation of the $^{252}$Cf coincidence counting rate provides the data to correct for the matrix around the plutonium inside the drum. Figure 3 shows a schematic diagram of the add-a-source method.

The measurement errors introduced from matrix materials in 200-L drums have been reduced by as much as an order of magnitude by use of this technique. In addition, this method can detect the presence of unexpected neutron shielding material inside the drum that might not allow the detection of nuclear materials.

For an in-plant installation[14] of the drum counter system, the detectability limit is 0.7 mg of $^{240}$Pu (or 2.1 mg plutonium) for a 15-min measurement. This excellent sensitivity was achieved using a special low-background detector design, good overhead shielding, and statistical techniques in the software to selectively reduce the cosmic-ray neutron background.

We are using Monte Carlo calculations to design high-efficiency (25-40%) waste drum counters so that the multiplicity counting technique can be combined with the add-a-source method. High efficiency is required to measure the triples counting rate with good statistical precision. For significant quantities of plutonium, the ratio of triples to doubles is proportional to the efficiency, including the matrix perturbation to the efficiency. This ratio technique also helps to correct for any localization or nonuniform distribution of the plutonium in the matrix. However, the statistical uncertainty in the triples rate is a problem for small plutonium masses.[15]

## 7. CONCLUSIONS

Passive neutron measurement systems have demonstrated excellent reliability and adaptability to a large range of inspection verification problems. The unattended, continuous-mode operation with automated data collection, storage, and convenient retrieval has made it possible for inspectors to spend less time in the plutonium facility without any loss of measurement capability. In fact, the sample constraints on size, mass, and containment dictated by the plant robotics system have made it possible to obtain a higher accuracy and precision with the NDA systems than has been possible for older conventional facilities. The precision and stability of the neutron systems is 0.1% to 0.2% and the accuracy depends on the fuel category.

With the installed NDA systems operating in a continuous mode, we obtain radiation data that gives a time history of the movement of nuclear materials in the facility. The integration of this continuous radiation data with the digital video data can effectively enhance the effectiveness of safeguards in facilities while reducing the inspector's time in the field. A new method for integrating the NDA data with the video data is presented in the paper "Safeguards Applications of Pattern Recognition and Neural Networks," presented at this meeting.[16]

The multiplication counting for impure samples is currently being evaluated for $^{244}$Cm measurements with applications to spent fuel and reprocessing plant safeguards. Additional work is required for the commercialization of the hardware, inspector-specific software, and inspector training related to multiplicity counting.

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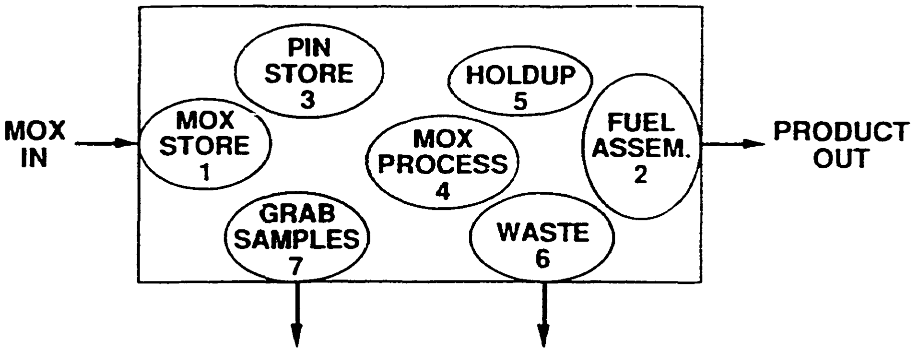
## FIGURE CAPTIONS

Fig. 1. Diagram of the PFPF facility MOX material locations and the corresponding NDA systems.

Fig. 2. Schematic diagram of the PSMC showing the location of the 80 helium 3 tubes and the graphite end plugs. The sample cavity height is 41 cm and the diameter is 20 cm.

Fig. 3. Conceptual diagram of the add-a-source configuration including the $4\pi$ $^3$He detector, the $CH_2$ shield for the $^{252}$Cf, and the nickel reflector block to scatter additional neutrons into the drum.



| Location | Detector System |
|---|---|
| 1 | PCAS - Canister input |
| 2 | FAAS - Fuel assembly output |
| 3 | FPAS - Fuel-pin counter |
| 4 | MAGB - Material accountancy glove-box counter |
| 5 | GBAS - Glove-box holdup counter |
| 6 | WDAS - Waste-drum assay system |
| 7 | INVS(Ge) - Inventory sample neutron and gamma assay |

Fig. 1

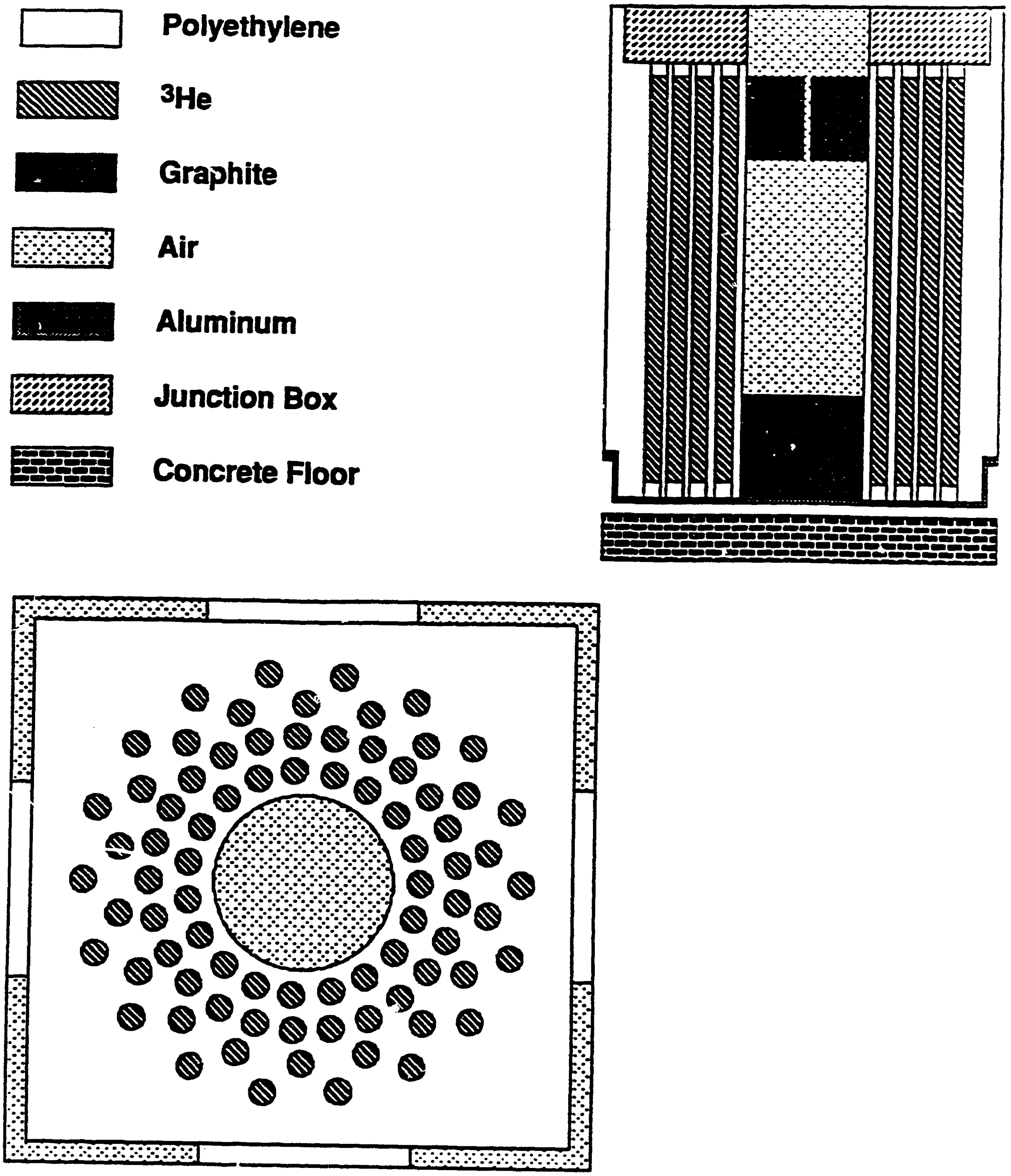

Polyethylene
3He
Graphite
Air
Aluminum
Junction Box
Concrete Floor

Fig. 2

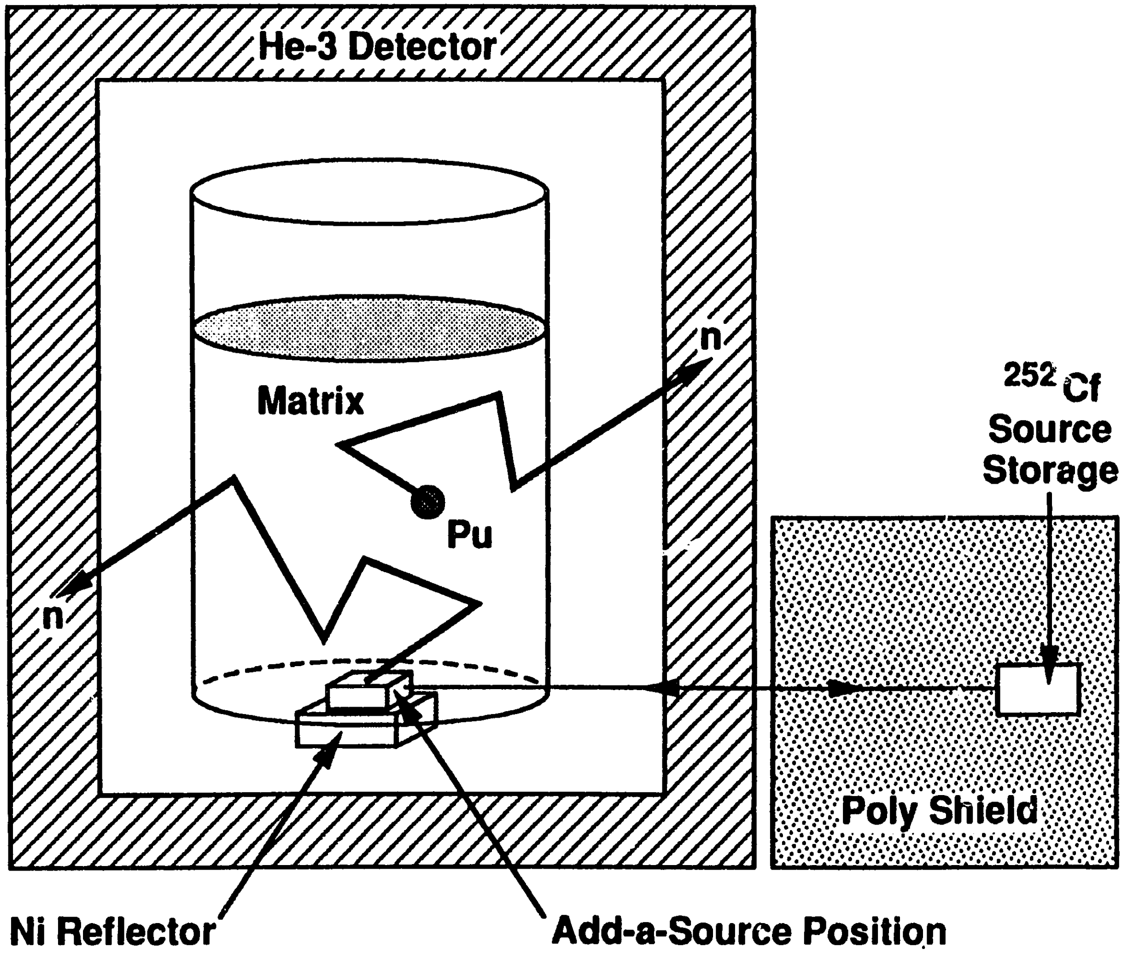

He-3 Detector
n
$^{252}$Cf
Source
Storage
Matrix
Pu
n
Poly Shield
Ni Reflector
Add-a-Source Position

Fig. 3

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