Passive Neutron Assay

Passive neutron assays detect the neutrons emitted when an isotope undergoes spontaneous fission. This is perhaps the easiest assay to perform. It is useful for measuring bulk materials because neutrons penetrate much farther than gamma rays. In practice, ^{240}Pu and ^{238}U are the only isotopes having sufficient spontaneous fission rates and isotopic abundances for passive neutron assay.

The assay is complicated by the fact that alpha particles from radioactive decays of uranium and plutonium interact with light elements in the matrix materials to produce additional neutrons. For example, if PuF_4 is present in the sample, neutrons will be produced through the reaction $^{19}F + \alpha \rightarrow {}^{22}Na + n$. However, each (α,n) reaction produces only one neutron in contrast to a spontaneous fission that produces a group of two or three. Consequently, we can differentiate spontaneous fission neutrons from the others by neutron coincidence counting. We have developed special time-correlation counting circuitry, called the shift register, to count coincidence neutrons and thus the spontaneous fission rate.

Neutron coincidence counting of ^{240}Pu spontaneous fission is the simplest method for the assay of plutonium provided the plutonium isotopic composition is known ahead of time. For high-burnup plutonium reactor fuels, a coincidence count may also contain a significant contribution from ^{242}Pu spontaneous fission. Since the spontaneous fission rate for ^{240}Pu is low, approximately 460 fissions/s per g ^{240}Pu, and the ^{240}Pu isotopic abundance is typically in the range 8-20%, a high-efficiency detection system is essential for rapid, quantitative assay. For uranium fuels, ^{238}U can be assayed by counting its spontaneous fissions, but the specific rate is much lower (approximately 7×10^{-3} fissions/s per g ^{238}U) and kilogram quantities are required. We have developed cylindrical detectors (with a central sample cavity) that have efficiencies as large as 20% for coincidence detection of fission events. These detectors are made of ^{3}He or BF_3 proportional counters and polyethylene. The sensitivity of the highest efficiency units is about 1 mg ^{240}Pu. Organic scintillation detectors have also been used for coincidence detection of spontaneous fission.

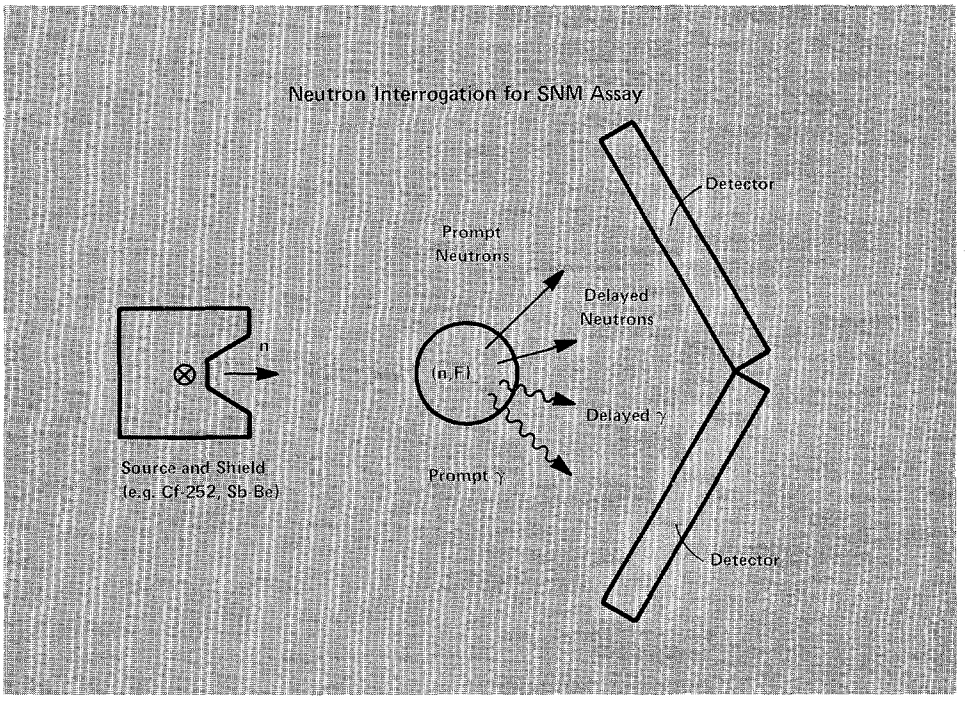

Fig. 15. The basic components of an active neutron assay system showing the interrogation source, the sample, and the detectors.

Active Neutron Interrogation

When the gamma-ray attenuation of materials is too high for passive assay, and the spontaneous fission rate is too low for neutron coincidence counting, we can interrogate the sample with highly penetrating beams of neutrons. The bombarding neutrons induce fissions in the sample and various gamma-ray and neutron signatures of the neutron-fission reactions are detected. During the past few years, active neutron interrogation has been used extensively to measure uranium and plutonium fuel rods, unirradiated fuel assemblies, and high-enriched uranium scrap. Either accelerator or radioisotopic neutrons could be used. Although accelerator neutron sources offer large yields and source modulation flexibility (that is, pulsing), they usually require a substantial amount of initial investment and technical support. On the other hand, radioactive neutron sources are reliable, simple, and less costly and are well suited for in-plant measurements and quality control of reactor fuel components as well as for some categories of scrap-material and process-stream measurements.

The basic components of an active neutron assay system are the neutron source, the sample, and the detectors to count the induced activities (Fig. 15). The neutron source is normally surrounded by materials to moderate or slow down the neutrons and tailor their energy for interrogation. These materials also are needed for personnel shielding. The fissions induced by the irradiation emit prompt and delayed neutrons as well as prompt and delayed gamma rays. One or more of these emitted signatures can be used for the assay depending on complexity of the measurement problem and the detection method.

Most fissionable material contains a mixture of both fissile and fertile isotopes. The fertile isotopes (for example, ^{238}U and ^{232}Th) have a threshold energy (about 1 MeV) for induced fis-

TABLE III
A NEUTRON IRRADIATION TECHNIQUE

Moderator Configuration[a] (Thickness in cm)	Total Leakage (%)	Median Energy (MeV)	Fraction of Neutrons With $E < {}^{238}U$ Threshold (%)	Fission Ratios[b] ($^{235}U/^{238}U$) Measured	Calculated
Pb: 7:5 (Cd)	146	1.76	39	003.19	003.10
W: 7:5 (Cd)	135	0.47	78	005.68	006.53
W/C: 7:5/7:5 (Cd)	122	0.19	79	015.3	013.3
$W/C/CH_2$: 7:5/7:5/2:5 (no Cd)	109	0.002	80	600.	788.

[a]The moderators and detectors were covered with 0.76-mm-thick cadmium where noted. DTF-IV calculations were used for the moderators with no cadmium; all others used the Monte Carlo code.

[b]Fission ratios are for equal weights of each fissionable isotope.

sion reactions, whereas fissile isotopes (for example, ^{235}U, ^{233}U, and ^{239}Pu) fission for all neutron energies. Active neutron interrogation techniques use subthreshold and superthreshold irradiation to separate the fissile and fertile components in a sample. The D-T, 14-MeV neutron generators are a convenient source of these interrogation neutrons, but the neutron energy is so high that both fissile and fertile isotopes undergo fission. To separate these components, we have developed neutron tailoring techniques to give both superthreshold and subthreshold irradiations from the same 14-MeV source. We have used Monte Carlo and DTF-IV neutron transport calculations to obtain maximum neutron leakage and desired neutron energy characteristics from various moderator assemblies. For example, for a 14-MeV neutron source, we have designed moderating assemblies that consist of concentric spherical shells of different materials including tungsten, graphite, and polyethylene. The large cross sections for the (n,2n) and (n,n′) reactions (incoming neutrons, outgoing neutrons) in tungsten reduce the energy of the 14-MeV source neutrons to a median energy of approximately 500 keV, and elastic scattering of neutrons in the graphite further reduces the average neutron energy to the low-keV region. Table III lists typical moderator configurations, where the notation $W/C/CH_2$: 7.5/7.5/2.5 indicates a 7.5-cm-thick core of tungsten surrounded by a 7.5-cm-thick shell of graphite surrounded by a 2.5-cm-thick shell of polyethylene. The total neutron leakage from the moderators exceeds 100% because of the large (n,2n) cross section in the heavy isotopes. Table III shows that the median neutron energy can be lowered from 14 MeV to a few keV using moderators with radii less than 18 cm. The tailoring assemblies given in Table III result in $^{235}U/^{238}U$ fission ratios from 3-600, a range that is very adequate for the separation of the fissile and fertile components in assay applications.

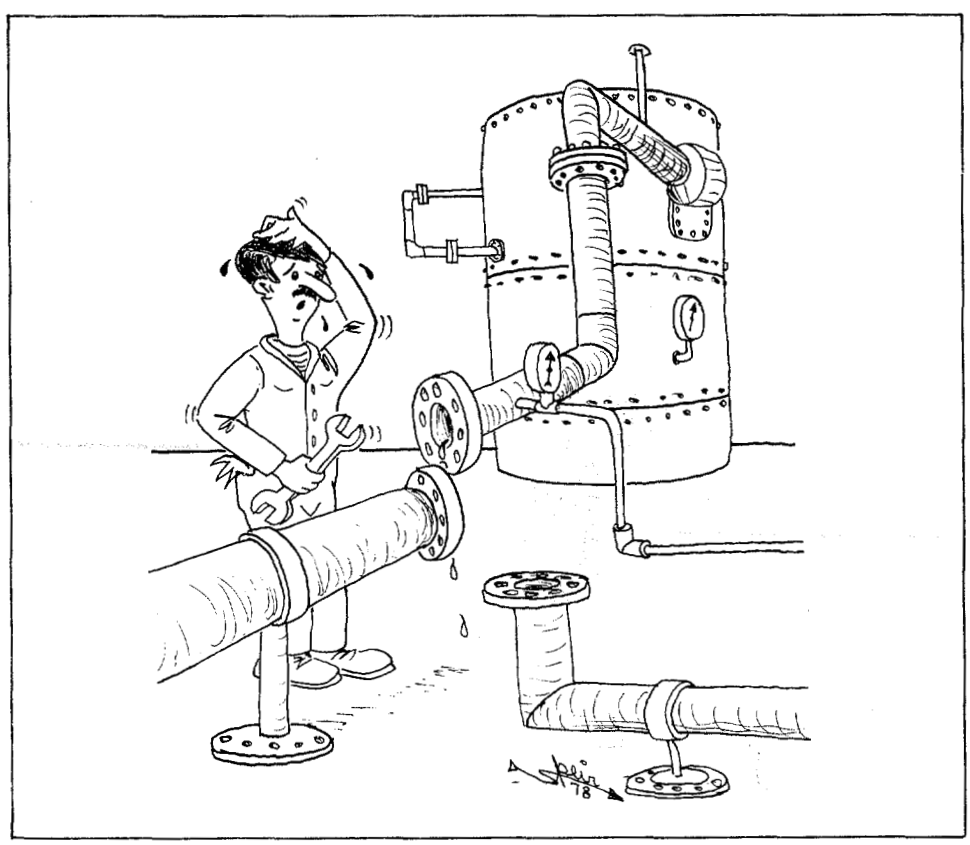

TABLE IV
RADIOACTIVE SOURCES FOR ACTIVE ASSAY APPLICATIONS

	Prompt n and γ		Delayed n and γ
Neutron Source	Integral	Coincidence	Integral
Sb-Be; 25 keV	n		
Ra-Be; 200 keV	n		
^{238}Pu-Li; 400 keV	n		
Am-Li; 400 keV		n and γ	
^{252}Cf; moderated		n and γ	n and γ
^{252}Cf; thermalized	n	n and γ	n and γ

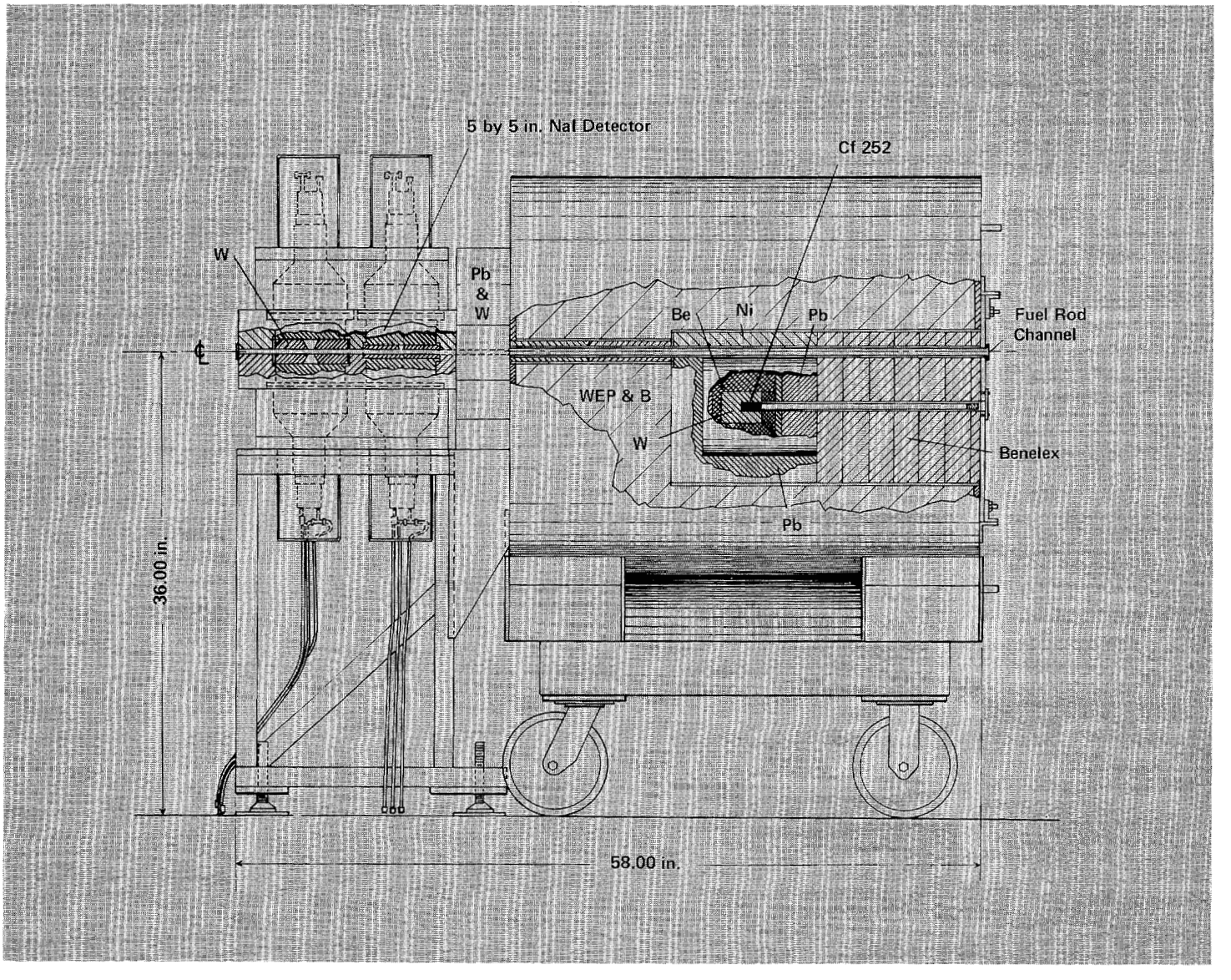

Fig. 16. Schematic of the ^{252}Cf fast-neutron assay system for the fast breeder reactor fuel rods. The delayed gamma rays induced by the fast-neutron irradiation are subsequently counted with the two NaI detectors, which also measure the passive gamma rays to detemine pellet-to-pellet uniformity.

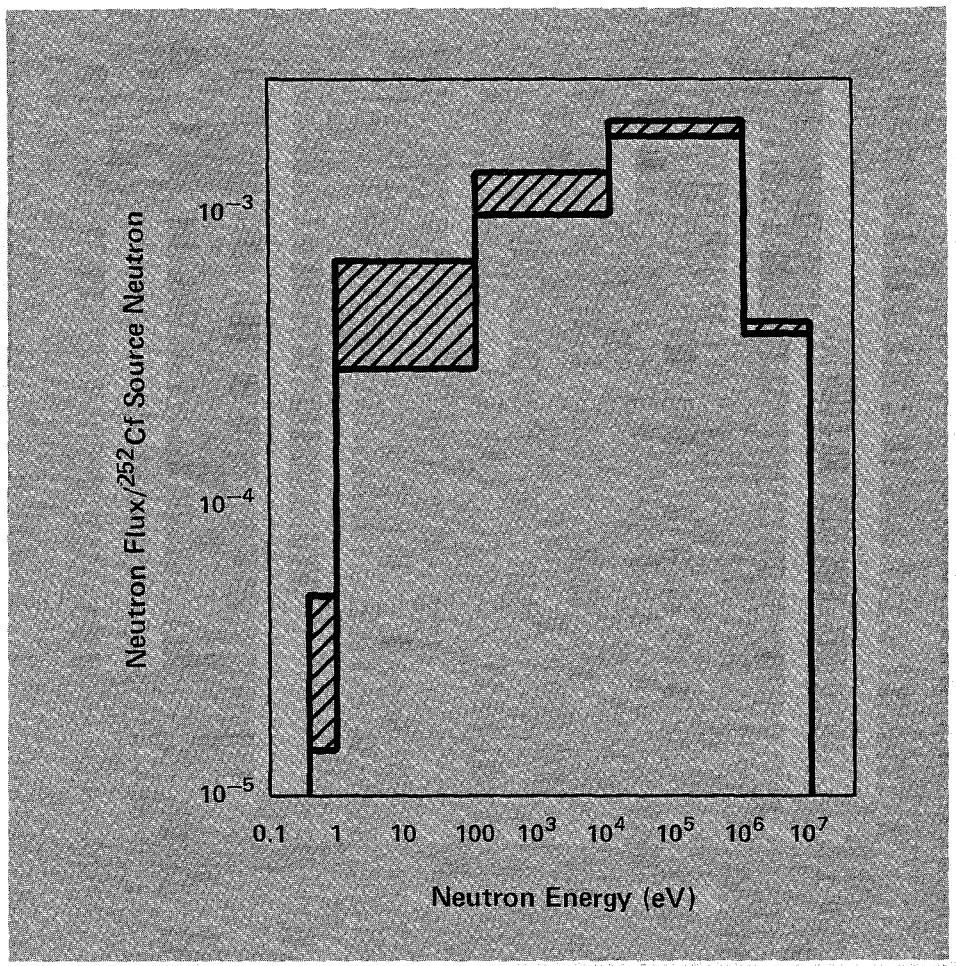

Fig. 17. Calculated neutron spectrum from a ^{252}Cf source for the system shown in Fig. 16 with (cross-hatched area) and without the cylindrical nickel fast-flux trap.

For most safeguards applications employing active neutron assay, radioactive sources have been preferred because of their simplicity and reliability.

To apply radioactive sources to a given measurement problem, three primary variables should be optimized: (1) the type of neutron source, (2) the moderator and shield for the source, and (3) the detector to count the induced neutrons and/or gamma rays. Radioactive neutron sources under investigation at LASL include ^{124}Sb-Be(γ,n), ^{88}Y-Be(γ,n), ^{226}Ra-Be(γ,n), and Am-Li(γ,n) (which have neutron energies primarily below the fission thresholds of ^{238}U and ^{232}Th), and ^{252}Cf (which emits higher energy neutrons).

Table IV gives a summary of radioactive sources and corresponding detection methods that have been used by LASL for assay applications. These applications range from portable field instrumentation used by inspectors to large in-plant installations used for waste and spent-fuel assay.

Fuel Rod Scanners

A high-performance instrument that incorporates several of the assay signatures previously described is the fuel rod scanner for the Fast Flux Test Facility (FFTF) at Hanford, Washington, a low-power reactor used to test designs for the Breeder Reactor Program. The design of the scanner required extensive use of Monte Carlo computer calculations to determine an optimum combination of materials, geometry, and detectors.

This hybrid assay system uses active neutron interrogation to determine total fissile content and passive gamma-ray spectroscopy to measure selected plutonium isotopes. The system measures the plutonium fissile content in a fuel rod to better than 0.5% accuracy and verifies that the plutonium content in individual fuel pellets within each rod is uniform from pellet to pellet. The fuel rods contain 70-80% natural uranium and 20-30% plutonium. The rods have an active length of 914 mm and a diameter of about 5 mm. Many thousands of these rods are used in the reactor core, and tight accountability of the plutonium is required for nuclear material safeguards.

A schematic of the system (Fig. 16) shows the 600-μg ^{252}Cf neutron source used for the interrogation, the moderator, the radiation shield, and the NaI detectors used to count the gamma rays. The neutron moderator is designed to give a fast-neutron irradiation (the irradiation channel is lined with cadmium) and a high fissile/fertile fission ratio (400/1). The fissile/fertile fission ratio corresponds to

$$\frac{\int \sigma^f_{239}(E)\,\phi(E)\,dE}{\int \sigma^f_{238}(E)\,\phi(E)\,dE}\,,\quad E > 0.4\ \text{eV}\,,$$

where σ^f_{239} and σ^f_{238} are the ^{239}Pu and ^{238}U fission cross sections, respectively, and $\phi(E)$ is the energy-dependent neutron flux in the irradiation channel.

Neutron transport calculations using both S_n and Monte Carlo techniques were used in the design of the ^{252}Cf source moderator. The calculations led to the design of the cylindrical moderator assembly shown in Fig. 16 with a tungsten core surrounded by a shell of beryllium followed by nickel and lead for fast-neutron reflection. The calculated neutron spectrum from a ^{252}Cf source for this system is shown in Fig. 17 with (cross-hatched area) and without the cylindrical nickel fast-flux trap. The presence of the nickel reflector increased the fission rate by approximately 70% over the lead reflector. This moderator resulted in a ^{239}Pu/^{238}U fission ratio of approximately 400/1 for irradiation neutrons above the cadmium cutoff energy (0.4 eV). It is necessary to keep this ratio high to avoid a significant contribution from ^{238}U fission, which is not of safeguards interest.

During operation, up to 30 fuel rods are placed in the loading magazine of the

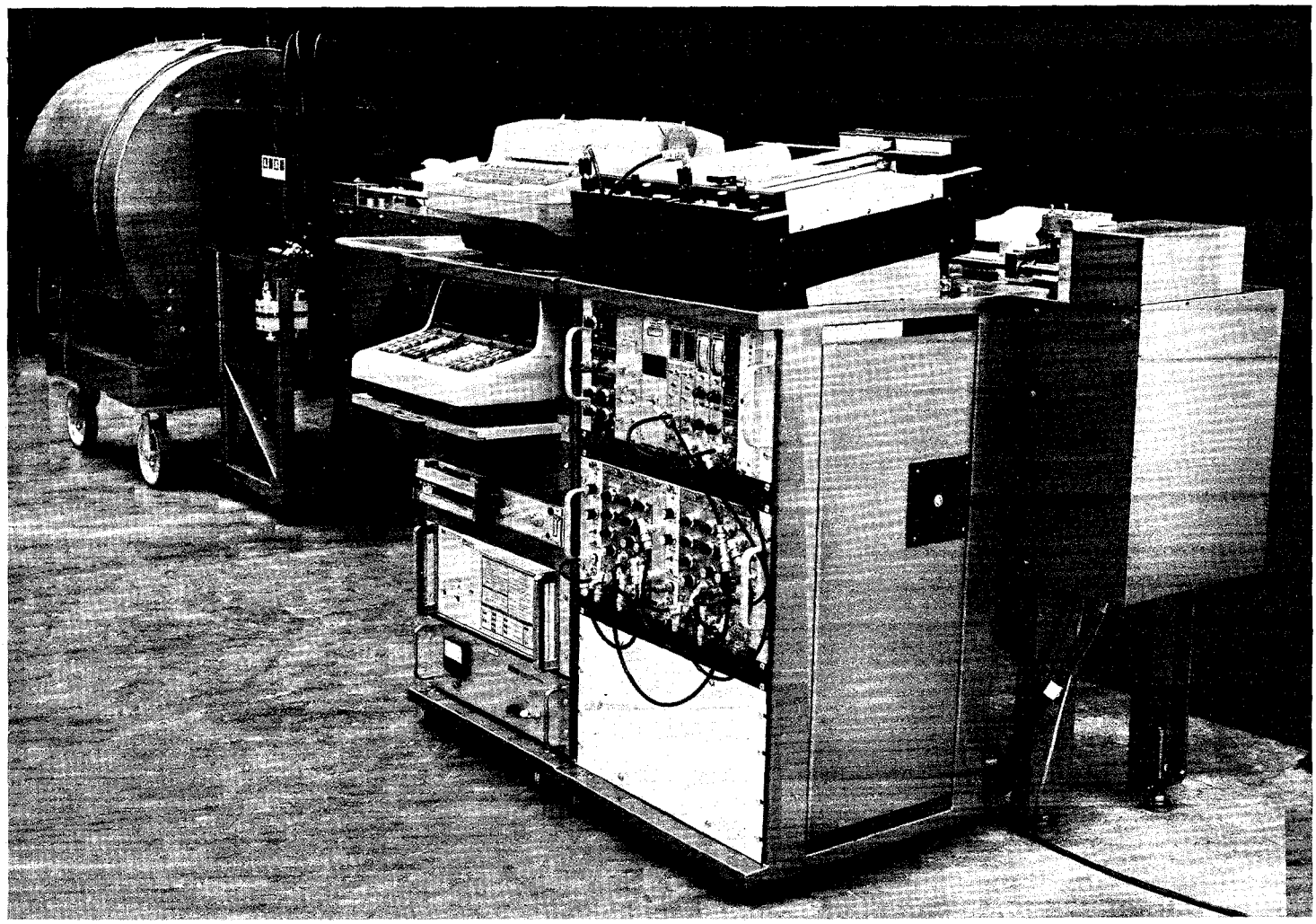

Fig. 18. Fast-neutron ^{252}Cf assay system for fast breeder reactor fuel rods. The system includes a 600-μg ^{252}Cf source and shield, two 5- by 5-in. NaI detectors to count the delayed gamma rays, automated fuel-rod handling, and a data-reduction system.

fuel-rod translator. The automated translator picks up the rod to be assayed and moves it through the NaI crystals to take a background count of the unirradiated rod and then into the ^{252}Cf assembly for irradiation with neutrons. The direction of travel is then reversed and the rod is withdrawn through the NaI detectors, which measure the delayed fission gamma rays for the total fissile assay and the passive gamma rays for the pellet-to-pellet uniformity. The two 5-in.-long by 5-in.-diam NaI detectors count the high-energy delayed gamma rays (>1200 keV) to determine the total fissile measurement because of their higher penetration through the rod. Simultaneously, the same NaI detectors determine pellet-to-pellet uniformity by measuring the much more intense lower energy passive gamma rays from the fuel. Each of the two NaI detectors examines a different low-energy window on a pellet-to-pellet basis, using a tungsten sleeve with a narrow collimation slit. One detector counts 60-keV gamma rays, which are mainly from ^{241}Am, and thus serves as a batch monitor because the ^{241}Am content of the fuel is time dependent. The second detector counts primarily plutonium gamma rays in the range of 100 to 500 keV. This second window provides a qualitative measure of the fissile plutonium in the rod. After the irradiation and scan of the fuel column, the rod is unloaded in the tray directly below the loading magazine and the cycle is repeated for the next rod.

The complete assay system, which includes automated fuel rod handling and computer-based data reduction, is shown in Fig. 18. This system was installed in 1973 at the Hanford Engineering Development Laboratory (HEDL) Plutonium Facility and has been used to measure the plutonium fissile content (better than 0.5% accuracy) and uniformity of over 70 000 FFTF fuel pins, about 40 000 of which are now being used in the reactor.

The ^{252}Cf-Based Assay System for the FAST Facility

We have designed a neutron interrogation assay system to measure waste solids from reprocessing and spent-fuel packages at the Fluorinel and Storage (FAST) Facility, a new addition to the Idaho Chemical Processing Plant used to reprocess spent fuels from the national defense program. Measurements of the canisters of solid waste will be performed for ^{235}U process control and accountability of the waste solids. The spent-fuel packages will be assayed to provide assurance that the ^{235}U content is below 10.5 kg, the criticality limit

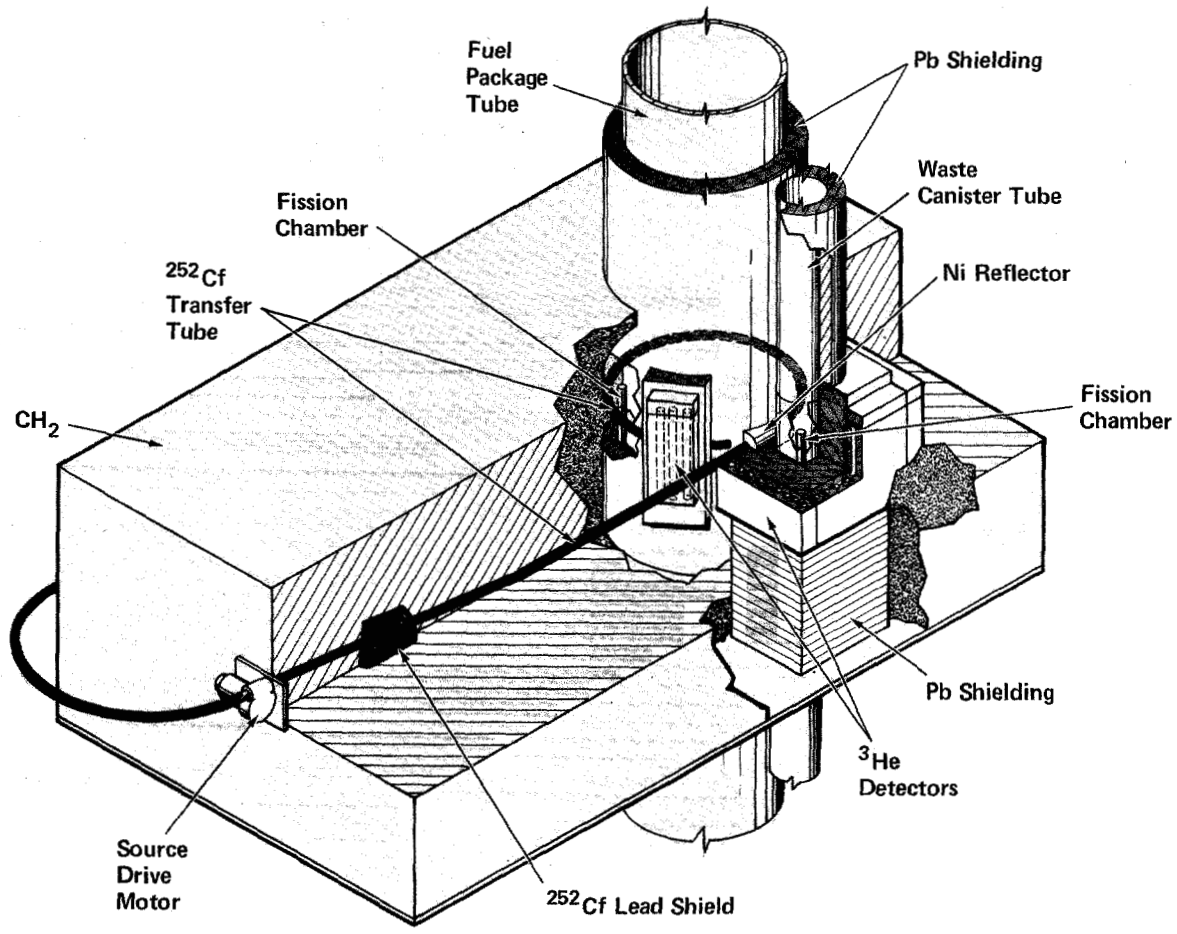

Fig. 19. The fast-neutron assay system for the Idaho Chemical Reprocessing FAST facility showing ^{252}Cf source transfer system, ^{3}He detectors, and the sample tubes for irradiated fuel elements and waste containers.

The assay system is based on the principle of the *shuffler*, which is a modulated ^{252}Cf source assay system that repetitively transfers the neutron source from the interrogation position to a shielded storage position while the delayed neutrons are counted. The assay system shown in Fig. 19 includes a source shield tank, a decoupling CH_2 shield, and an irradiation area. The assay sample is irradiated in a chamber surrounded by ^{3}He neutron detectors that are used to count delayed neutrons produced following each irradiation. We have evaluated the shuffler system for the assay of fuel rods, inventory samples, scrap and waste, uranium ore, irradiated fuel, and plutonium mixed-oxide fuel.

At the FAST Facility, the fissile contents vary from 0 to 400 g in the waste and from about 8 to 12 kg in the spent-fuel elements. Measurements must be obtained in the presence of large neutron (1.2×10^7 n/s) and gamma (50 000 R/h) backgrounds. The system employs fast-neutron interrogation of the sample using a large (5-mg) ^{252}Cf source to override the neutron backgrounds. Preliminary Monte Carlo calculations indicate that measurement precisions of better than $\pm 5\%$ ($2\text{-}\sigma$) are obtainable for waste loadings in excess of 200 g ^{235}U.

We are designing the assay instrument as an integral part of the FAST Facility with close cooperation from Exxon and Ralph M. Parsons Company (Architectural Engineer). The facility will house the assay instrument in a separate cubicle connected by through tubes to the fluorinel dissolution cell. Waste canisters and spent-fuel packages will be lowered by crane into the assay instrument through the tubes that penetrate the cell floor and extend below the assay system. The assay system will be remotely controlled and operated by a dedicated mini-computer system. Design of the instrument as an integral part of the facility, rather than as a retrofit, enables both facility operations and assay measurements to be better coordinated.

The Active Well Coincidence Counter (AWCC) for Portable Inspection Applications

An important safeguards area that has very different requirements from in-