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# NEUTRON ACTIVATION ANALYSIS AT THE CALIFORNIUM USER FACILITY FOR NEUTRON SCIENCE

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# Neutron Activation Analysis at the Californium User Facility for Neutron Science

**R. C. Martin, E. H. Smith, D. C. Glasgow, E. A. Jerde, D. L. Marsh, and L. Zhao**

## Abstract

The Californium User Facility for Neutron Science has been established to provide $^{252}$Cf-based neutron irradiation services and research capabilities including neutron activation analysis (NAA). A major advantage of the CUF is its accessibility and controlled experimental conditions compared with those of a reactor environment. The CUF maintains the world's largest inventory of compact $^{252}$Cf neutron sources. Neutron source intensities of $\leq 10^{11}$ neutrons/s are available for irradiations within a contamination-free hot cell, capable of providing thermal and fast neutron fluxes exceeding $10^8$ cm$^{-2}$ s$^{-1}$ at the sample. Total flux of $\geq 10^9$ cm$^{-2}$ s$^{-1}$ is feasible for large-volume irradiation rabbits within the $^{252}$Cf storage pool. Neutron and gamma transport calculations have been performed using the Monte Carlo transport code MCNP to estimate irradiation fluxes available for sample activation within the hot cell and storage pool and to design and optimize a prompt gamma NAA (PGNAA) configuration for large sample volumes. Confirmatory NAA irradiations have been performed within the pool. Gamma spectroscopy capabilities including PGNAA are being established within the CUF for sample analysis.

## 1. INTRODUCTION

This paper describes ongoing activities and future plans for neutron irradiations, PGNAA, and conventional instrumental neutron activation analysis (INAA) at the Californium User Facility for Neutron Science (CUF) at Oak Ridge National Laboratory (ORNL). The heavy isotope $^{252}$Cf is an intense neutron emitter that can be conveniently encapsulated in compact, portable neutron sources.[1] A single small source can contain up to 50 mg of $^{252}$Cf (~$10^{11}$ neutrons/s). When a nuclear reactor is impractical or too expensive, $^{252}$Cf can serve as an alternative source of neutrons for lower flux applications. Californium-252 has a half-life of 2.645 years and decays by alpha emission and spontaneous fission, with one milligram of $^{252}$Cf generating $2.314 \times 10^9$ neutrons/s (average energy of 2.1 MeV, most probable energy of 0.7 MeV). Common applications for $^{252}$Cf-based NAA include INAA for trace multielemental analysis and PGNAA for rapid nondestructive assay of the principal elemental contents of a sample (e.g., coal, cement, explosives, and chemical munitions). Other common applications of $^{252}$Cf include neutron radiography (e.g., aircraft and weapons components among others), fissile material and transuranic waste analyses, fuel rod scanning for enrichment and uniformity, start-up sources for nuclear reactors, and cancer therapy.

The Radiochemical Engineering Development Center (REDC) at ORNL processes the national supply of heavy elements after production at the neighboring High Flux Isotope Reactor (HFIR). The REDC Californium Facility purifies, encapsulates, and supplies much of the world with $^{252}$Cf and stores the national inventory of $^{252}$Cf sources for the U.S. Department of Energy (USDOE) Californium Sales/Loan Program, which includes university, government, and research loans. The CUF was established in 1996 to make $^{252}$Cf sources and portions of the Californium Facility available to outside researchers and to promote cost-effective research into the applications of $^{252}$Cf. Sealed sources containing up to 50 mg of $^{252}$Cf can routinely be used within one uncontaminated, walk-in hot cell, and larger masses of $^{252}$Cf can be used for irradiations within the water-filled $^{252}$Cf storage pool. Previous and potential applications of the CUF include thermal, fast, and prompt gamma NAA, neutron degradation of radiation detectors, dosimetry and cell killing experiments for neutron tumor therapy, and neutron radiography. The CUF is ideal for experiments requiring moderate neutron fluxes such as radiation effects on electronic systems.

The CUF is developing capabilities for both INAA and PGNAA. Several experimental configurations are available for high-flux NAA of samples. In-cell, samples can be placed adjacent to high-intensity $^{252}$Cf sources within a source-moderator module for NAA. In the $^{252}$Cf storage pool, larger $^{252}$Cf masses (up to hundreds of milligrams) can be used for irradiations by inserting a sample-containing pneumatic rabbit adjacent to several source-containing rabbits. The achievable neutron fluxes in these in-cell and in-pool configurations were estimated using the MCNP-4B Monte Carlo neutron transport code. Dosimetric measurements were obtained for the in-pool configuration. MCNP-4A was used to develop an optimized $^{252}$Cf source–moderator–sample–detector and shielding design for PGNAA of large

volume samples such as coal analysis. The design mass and footprint was then reduced for in-cell installation. Finally, a design is being developed for a $^{252}$Cf-based pneumatic transfer and INAA system with operation planned for 1999. As planned, this system would use the largest mass of $^{252}$Cf (nominally 200 mg) ever assembled for a single application.

# 2. BACKGROUND

## 2.1 CALIFORNIUM-BASED INAA SYSTEMS

An early application of $^{252}$Cf-based INAA involved the analysis of moon rocks at the University of Kentucky for the National Aeronautics and Space Administration.[2] The largest $^{252}$Cf INAA system to date was established in 1976 at the Savannah River Technology Center of Savannah River Laboratory (SRL) and employed a maximum of 100 mg of $^{252}$Cf. With a peak thermal neutron flux of $2 \times 10^9$ cm$^{-2}$ s$^{-1}$, this facility provided analysis for solid and liquid samples such as alloys, sediments, rocks, and site process solutions. Most convenient was the capability for analysis of bulk samples without significant sample preparation. Over 40 elements could be detected at the sub-ppm level, with precisions and accuracies of ±10% reported for most elements.[3] The pneumatic transfer system with its 13-mL high-density polyethylene (HDPE) rabbits permitted analysis of short-lived activation products including capabilities for automated cyclic irradiations.[4] Another high-flux $^{252}$Cf INAA system established by the Oak Ridge Associated Universities in Oak Ridge, Tennessee was primarily used for the environmental analysis of contaminants in soils, sediments, and sludges.[5] Forty milligrams of $^{252}$Cf in a graphite moderator assembly provided a thermal neutron flux of $2 \times 10^8$ cm$^{-2}$ s$^{-1}$, sufficient to analyze over 40 elements and to detect parts-per-million levels of contaminants such as uranium, thorium, chromium, mercury, silver, and arsenic.

## 2.2 PGNAA SYSTEMS

While INAA continues to be an important application of $^{252}$Cf sources, the primary commercial use of $^{252}$Cf is in the application of PGNAA to process control in the coal and cement industries, among others. Typical source sizes range from 5 to 100 $\mu$g of $^{252}$Cf, depending on the desired sensitivity. The intensities of the prompt gamma ray emissions are primarily a function of the elemental composition of the sample and the size of the $^{252}$Cf source, plus matrix effects such as bulk density, porosity, and moisture content. Elements commonly detected by PGNAA include chlorine, sulfur, phosphorus, nitrogen, arsenic, iron, and hydrogen. Commercial coal analyzers which measure sulfur, hydrogen, and ash are used for on-line sorting and blending of coal to control atmospheric emissions. A comparable system is used for cement analyses. $^{252}$Cf sources have also been applied to the detection of explosives and other chemical munitions by PGNAA, from a first-generation airport luggage inspection system developed and installed by Science Applications International Corporation[6] to the system recently marketed by EG&G ORTEC for nondestructive analysis of artillery shells and other ordinance containers.[7] This Portable Isotopic Neutron-Spectroscopy (PINS) Chemical Assay System can distinguish between high explosives, nerve agents, poison gases, and other chemical components in unexploded military ordinance.

## 2.3 IRRADIATION CAPABILITIES OF THE CALIFORNIUM USER FACILITY

A major advantage of the CUF is its accessibility and controlled experimental conditions compared with those of a reactor environment. For example, neutron damage effects are predominant in $^{252}$Cf irradiations, with ~10:1 neutron:gamma dose equivalent ratio. The CUF encompasses two uncontaminated walk-in hot cells, the water-filled $^{252}$Cf storage pool, available neutron sources, pneumatic transfer capabilities, and associated infrastructure and technical support. After hands-on experimental setup inside the hot cell, all personnel are evacuated, the cell is closed, and $^{252}$Cf sources are pneumatically transferred into the cell and positioned remotely using manipulators. Individual source strengths from <1 $\mu$g to tens of milligrams of $^{252}$Cf are available, with total strengths up to 50 mg (~$10^{11}$ neutrons/s) permitted in one cell and 5 mg in the other. The storage pool is a large water tank containing the CUF inventory of sealed $^{252}$Cf sources (typically up to several hundred milligrams of $^{252}$Cf). For pool irradiation, a sample can either be pneumatically transferred into the storage lattice and the neutron field tailored by rearranging the neighboring sources, or larger samples and watertight experiments can be physically lowered into the underwater neutron field. For convenience in performing multiple sequential irradiations, samples up to 8.5 cm in diameter can be transferred into the closed hot cell using a sample port, and irradiated samples less than 5.7 cm in diameter and without transferrable contamination can be removed from the closed hot cell using a sample pull drawer.

### 2.3.1 Neutron Damage Testing of Semiconductor Devices

An ongoing project demonstrates neutron damage testing of electronic devices. To test the radiation hardness of a variety of avalanche photodiode detectors to be used in high-energy physics experiments, a team from Northeastern University and the University of Minnesota assembled an experiment in which 6 to 48 mg of $^{252}Cf$ was used to approximate the expected in-service neutron flux and fluence at the Compact Muon Solenoid (CMS) experiment at CERN.[8] Real-time data acquisition and computer control of the in-cell experiment were provided by cables to the out-of-cell hardware. The experiments simulated the expected in-service fast neutron flux of $2 \times 10^5$ cm$^{-2}$ s$^{-1}$, and accelerated irradiations provided a fast fluence which approximated that expected from five years of CMS operation; i.e., up to $1.2 \times 10^{13}$ cm$^{-2}$ for a 335 hour exposure.

### 2.3.2 Irradiation of Cancer Cells for Boron Neutron Capture Therapy

Novel boron-containing compounds are synthesized at the University of Tennessee, Knoxville. To test their relative effectiveness for cancer treatments employing boron neutron capture therapy (BNCT), a simple irradiation experiment was conducted in which living lung cancer cells impregnated with the boron compounds were exposed to $^{252}Cf$ neutrons for periods of 0.5 to 4.0 minutes.[9] The relative cell survival fractions after irradiation indicate the efficacy of each compound for use in BNCT. The schematic of the simple polyethylene irradiator is shown in Figure 1a. Four sources like that shown in Figure 1b and containing a total of 28.1 mg of $^{252}Cf$ were arranged symmetrically within the twelve outer holes of the sample irradiator. The central compartment was filled with water, and the irradiator cap positioned the 1.5-cm diameter test tube containing the cells in the center of the irradiator. The centerline distance of 3.0 cm between source and sample included 0.95 cm of polyethylene and 1.3 cm of water. This configuration provided a thermal neutron flux of $2 \times 10^8$ cm$^{-2}$ s$^{-1}$ at the sample as measured by activation of manganese wire. Addition of more moderator material around the outer surfaces of the irradiator would have increased this thermal flux. No attempt was made to optimize the thermal-to-fast neutron flux ratio.

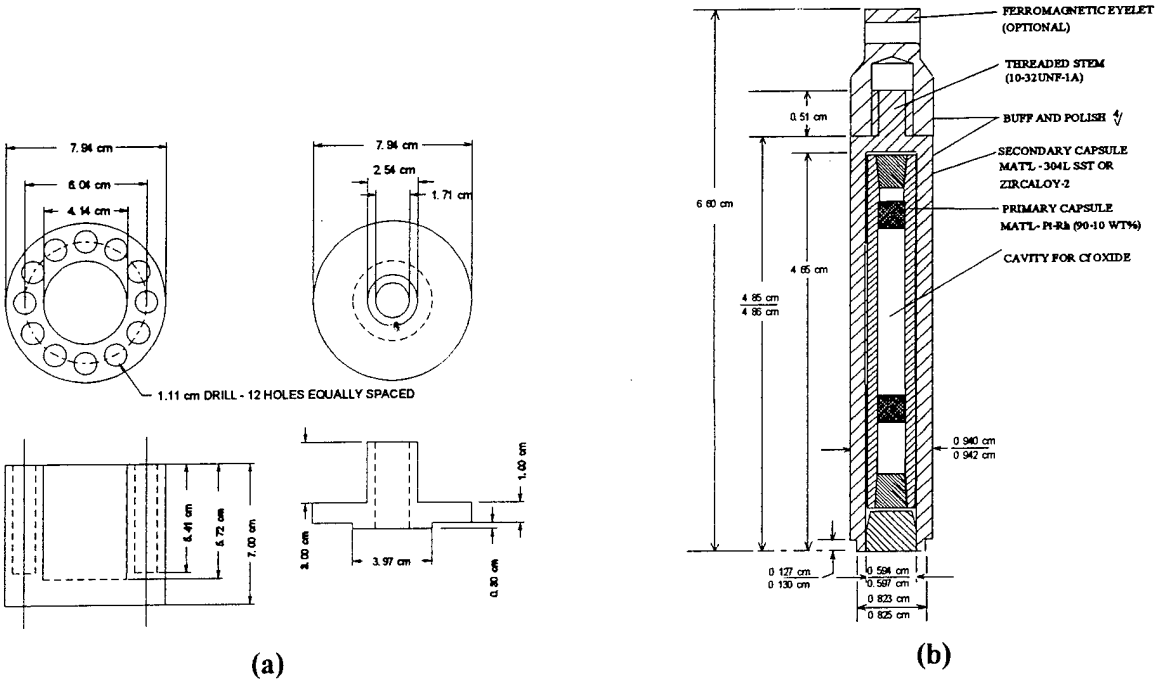


**Fig. 1. Experimental configuration for BNCT experiments: (a) Neutron source holder with cap to center the test tube; (b) SR-Cf-3000 series $^{252}Cf$ source design, used for BNCT and most high-flux irradiations.**

# 3. DESCRIPTION

## 3.1 IN-CELL NAA

Up to 50 mg of $^{252}Cf$ can be used for a single application within one hot cell, and more can be used if the irradiation is confined within a $^{252}Cf$ shipping container inside the cell. Three different irradiation configurations are presently available in cell, two with different HDPE moderator designs and a third contained within a shipping cask. In the first, a modular HDPE moderator block of dimensions 97.2 cm width × 40.6 cm depth × 45.7 cm height, originally designed for PGNAA applications, was configured for irradiation of samples up to 3.8 cm in diameter. Three holes were drilled down into the middle of the block, at centerline distances of 6.8, 13.6, and 20.3 cm from the vertical block surface into its 40.6 cm depth. Either samples or $^{252}Cf$ sources can be inserted into any of the holes. MCNP-4B was used to estimate the thermal and fast neutron fluxes at the sample holes from 50 mg of $^{252}Cf$.

A prototype sample irradiator design was shown in Figure 1a for the BNCT experiments. This design was duplicated in a second irradiation configuration with more polyethylene around the cylindrical irradiator module to increase the thermal flux achievable for a given $^{252}Cf$ mass. Compared to Figure 1a, the diameter of the module was increased from 7.9 to 9.8 cm and the height was increased from 7.0 to 10.8 cm to increase polyethylene mass at the radial and bottom surfaces. The 12-source concentric irradiator design around a sample cavity of 4.1 cm diameter by 5.7 cm height was duplicated. For greater thermal flux, this module can be positioned inside a central hole within HDPE blocks of dimensions 26.0 cm × 26.0 cm, another polyethylene cylinder can be placed on top of the module, and the entire unit placed on polyethylene slabs. No MCNP modelling of this configuration was performed.

The third in-cell configuration, irradiation within a $^{252}Cf$ shipping cask placed inside the hot cell, is not limited to the $^{252}Cf$ 50 mg limit. The previous 12-source irradiator module was fabricated to fit inside a large shipping cask which can ship 60 to 80 mg of $^{252}Cf$. Placement of two of these irradiation modules within the cask will permit sample irradiations with a maximum of 100 mg of $^{252}Cf$. Computations have not been performed to estimate the maximum neutron flux for this configuration.

## 3.2 IN-POOL NAA

The $^{252}Cf$ storage lattice is portrayed in Figure 2, looking down into the pool. The lattice tubes are made from 6061-T6 aluminum alloy. Up to three large pneumatic rabbits can be placed in each lattice position. Each rabbit has internal dimensions of 11.8 cm length × 3.6 cm diameter and can hold multiple $^{252}Cf$ sources. The screw cap has an O-ring to prevent water ingress. The source-containing rabbits can be rearranged within the lattice at will to tailor the neutron flux and energy. Maximum flux is obtained by positioning high-intensity $^{252}Cf$ sources on four sides of the lattice tube with the sample-containing rabbit. Rabbits are pneumatically transferred between the hot cell and the storage pool, then manually lowered or removed from the storage lattice. An irradiated sample can be transferred from the storage pool, opened, and removed from the cell within minutes. An MCNP-4B input file was created to model the entire storage lattice, and calculations performed to estimate the flux available at a sample position. Several irradiations, some with flux monitors, were performed to demonstrate the irradiation procedures and compare to computational results. The pool typically contains less than 100 mg of $^{252}Cf$, but after a heavy element processing campaign an additional 300 mg or more of $^{252}Cf$ can be made available.

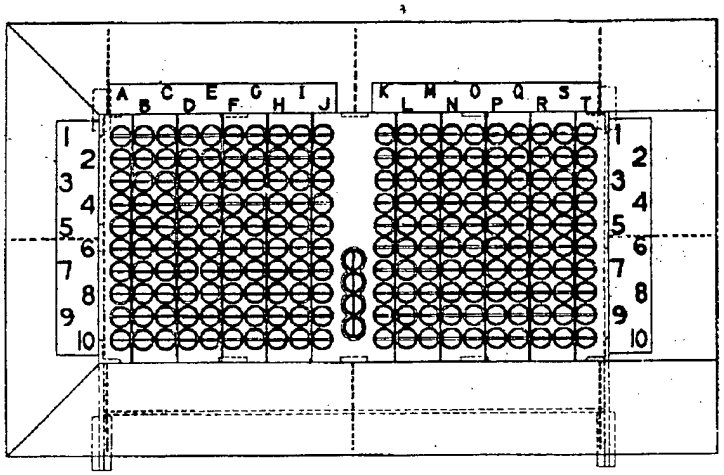


**Fig. 2. Layout of the storage lattice within the water-filled $^{252}Cf$ storage pool (top view).**

## 3.3 AUTOMATED INAA RABBIT SYSTEM

The Food and Drug Administration (FDA) would like to establish a permanent $^{252}$Cf-based INAA facility for analyzing food samples. ORNL staff have suggested this facility be established as part of the CUF. As the national storehouse for the USDOE Californium-252 Program, the CUF has unique infrastructure for installing a very cost effective facility. Based on four 50-mg $^{252}$Cf sources, this activation facility would provide a unique resource for the radiochemical analysis of samples and demonstration of $^{252}$Cf capabilities for INAA using more $^{252}$Cf than any other facility in the world, past or present. The activation facility would also provide a versatile complement to the INAA Laboratory at HFIR, expanding the range of samples amenable to INAA and providing continuous backup during extended HFIR outages. Planning for this system is ongoing between the REDC and the FDA, in collaboration with the University of Tennessee, Knoxville and with the Chemical and Analytical Sciences Division of ORNL. The goal is operational status in late 1999, prior to an extended HFIR maintenance outage to provide continuous INAA services to the ORNL and user community.

The sample irradiation module will be located within the $^{252}$Cf storage pool. An existing pneumatic sample transfer and control system, which was previously fabricated for FDA use, will couple the irradiation module to the assay station on the floor above the pool. The available $^{252}$Cf stockpile at the CUF can be used to augment the dedicated FDA sources for higher sensitivity or for neutron damage studies. Although $^{252}$Cf cannot match HFIR's neutron fluxes, the $^{252}$Cf sources are never subject to reactor downtime, temperatures remain near room temperature, and the "coolant" is not pressurized. The unlimited irradiation times and use of large 40-mL sample rabbits partially offset the lower neutron flux. The large rabbit volumes permit use of neutron-absorbing sleeves for spectral tailoring (epithermal and fast NAA) to maximize sensitivities for specific elements. Gamma heating from $^{252}$Cf-based activation is insignificant, permitting irradiation of liquid samples as well as volatile and delicate samples such as biological specimens.

## 3.4 PGNAA

For PGNAA, the MCNP-4A Monte Carlo neutron transport code was used to develop an optimized $^{252}$Cf source–moderator–sample–detector and shielding design for large volume samples such as coal analysis. The design mass and footprint was reduced for in-cell installation and modified for smaller sample geometries. PGNAA capabilities were tested with a gamma spectroscopy system consisting of a 5" x 5" NaI detector in-cell coupled to an EG&G ORTEC 456 high-voltage power supply, ORTEC 113 preamplifier, Canberra 2025 spectroscopy amplifier, and ACCUSPEC-A data acquisition board out-of-cell. Over 8 meters of cable were required to couple the electronics to the detector.

The large-sample PGNAA system was designed via a series of calculations to optimize the thermal-to-fast neutron flux ratio at the sample position for each component of the PGNAA system. Details and numerical results will be presented elsewhere, but the computational methodology will be outlined here. The goal of this effort was an optimized system for coal analysis, although the same principles and design can be applied to the general analysis of stationary and moving bulk samples. The dimensions of a rectangular bulk moderator block consisting of HDPE were first established to optimize the thermal-to-fast flux ratio. The sample channel was positioned along one face of the bulk moderator (see Figure 3). The $^{252}$Cf source was positioned inside a smaller moderator block which was inserted into the bulk moderator along its bottom surface. The dimensions of the smaller moderator block were optimized, followed by selection of distance between the $^{252}$Cf source and the sample channel. A variety of materials were evaluated for the smaller moderator block (polyethylene, polyethylene with 0.5% boron, $D_2O$, $H_2O$, BeO, and liquid hydrogen). Although the liquid hydrogen provided the largest flux ratio, polyethylene was chosen for overall convenience. The width of the sample channel was chosen. The angle of the detector axis in relation to the sample channel axis was evaluated for 0°, 45°, and 90°, with 45° chosen to minimize neutron flux on the detector. The detector was positioned across the sample channel from the source and moderator. Lithium-6 hydride was preferred over polyethylene-5% boron to shield the detector from neutrons, and the detector shielding was enclosed in a bismuth-lead alloy box. For a given $^{252}$Cf source strength and detector size, the neutron and photon flux at the detector face was calculated as a function of detector distance from sample channel. Finally the thermal-to-fast neutron flux ratio was averaged across the sample channel and determined at the channel axis.

This computational process determined the optimum system design for a large sample (channel dimensions of 121 cm × 41 cm × 20.5 cm), but the total system as designed would weigh several tons. Therefore, another design iteration reduced the system footprint and total mass for in-cell installation. This final design is shown in Figure 3. The

central sample channel is bracketed on one side by the source moderator (the three holes provide choice of source position) and on the other by more HDPE moderator and the NaI detector surrounded by lead bricks.

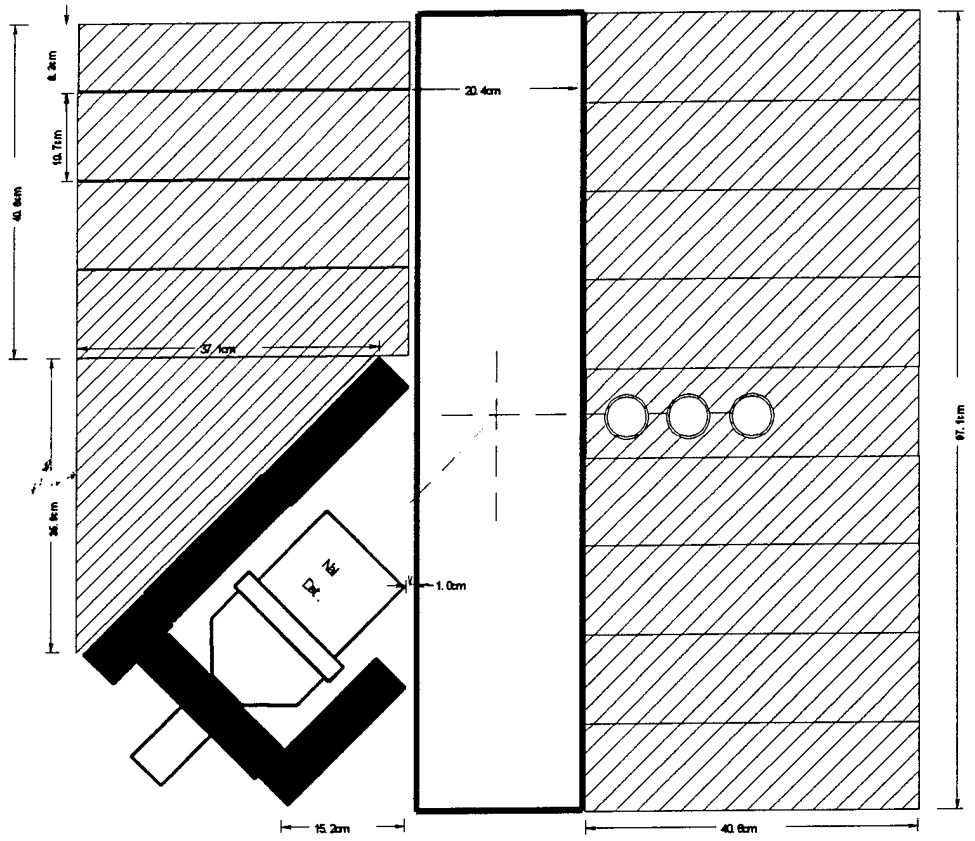


**Fig. 3. Optimized in-cell PGNAA system design for the analysis of bulk samples.**

# 4. RESULTS

## 4.1 IN-CELL IRRADIATION CONFIGURATIONS

As previously mentioned, the thermal flux measured for the BNCT experiment was $2 \times 10^8$ cm$^{-2}$ s$^{-1}$, using 28.1 mg of $^{252}$Cf and moderated by 2.25 cm of polyethylene and water.

For MCNP-4B modelling of the large polyethylene block moderator with three source-sample holes drilled down to the middle of the block, a neutron energy of 0.414 eV was used to differentiate between thermal and fast neutrons. For 50 mg of $^{252}$Cf in the center of the moderator block, the thermal and fast fluxes at the neighboring hole (i.e., at a centerline distance of 6.8 cm) were $5.9 \times 10^8$ and $2.7 \times 10^8$ cm$^{-2}$ s$^{-1}$ respectively (thermal-to-fast ratio of 2.2), and in the hole toward the outer surface (centerline distance of 13.5 cm) the fluxes were $1.1 \times 10^8$ and $2.5 \times 10^7$ cm$^{-2}$ s$^{-1}$ (thermal-to-fast ratio of 4.4). Calculations were performed for $10^7$ particle histories. The thermal flux results for the neighboring hole are within a factor of two of the design calculations presented by Nichols.[10] No flux advantage was gained by placing the sample in the middle hole and dividing the $^{252}$Cf into 25 mg in each of the two adjacent holes: the thermal flux was again $5.9 \times 10^8$ and the fast $2.6 \times 10^8$ cm$^{-2}$ s$^{-1}$.

No MCNP modelling of the second polyethylene moderator configuration (i.e., the enhanced BNCT experiment configuration) was performed, but compared to the BNCT experiments and the previous modelling for the larger polyethylene block irradiator, a thermal flux > $5 \times 10^8$ cm$^{-2}$ s$^{-1}$ and total flux ~ $10^9$ cm$^{-2}$ s$^{-1}$ at the sample position could be expected from 50 mg of $^{252}$Cf. This projection for thermal flux is consistent within a factor of two of Nichols' results.[10] No modelling of the neutron flux within the cask configuration was performed. A maximum thermal flux approaching$10^9$ cm$^{-2}$ s$^{-1}$ could be expected with the use of 100 mg of $^{252}$Cf.

## 4.2 STORAGE POOL IRRADIATONS

The five pneumatic storage rabbits containing the largest $^{252}$Cf masses available at the time were selected and arranged around a vacant lattice position. Each of these rabbits contained several high-intensity SR-Cf-3000 series sources (Figure 1b). Referring to the storage lattice portrayed in Figure 2, a rabbit containing 32.7 mg of $^{252}$Cf was placed into lattice position O3, a rabbit with 32.4 mg into Q3, a rabbit with 20.1 mg into P4, a rabbit with 9.3 mg into P2, and a rabbit with 4.3 mg into Q2. Central lattice position P3 was reserved for the samples. In total, 4 rabbits containing 94.5 mg of $^{252}$Cf were positioned as nearest neighbors to the irradiation position (at a centerline distance of 6.35 cm) and the fifth rabbit with 4.3 mg as next-nearest neighbor at 9.0 cm. Of the 6.35-cm distance, approximately 0.5 cm consists of aluminum alloy (lattice tube and rabbit walls) and 2.0 cm of water moderator.

The MCNP-4B calculations were performed with $10^6$ particle histories to estimate the thermal (energy <0.414 eV) and fast neutron flux at a vacant (water-filled) lattice position with 50 mg of $^{252}$Cf in a nearest-neighbor position. The total neutron flux was calculated to be $8.2 \times 10^8$ cm$^{-2}$ s$^{-1}$ and thermal flux of $4.5 \times 10^8$ cm$^{-2}$ s$^{-1}$, giving a thermal-to-fast flux ratio of approximately 1.2. This thermal flux is again within a factor of two of Nichols' design calculations.[10] Normalizing this result to the 98.8 mg of $^{252}$Cf actually used, an experimental thermal flux at the sample position could be expected in the range of $8.9 \times 10^8$ cm$^{-2}$ s$^{-1}$ (disregarding the small error from one rabbit in the next-nearest neighbor position). With a rabbit positioned in the irradiation position, displacing water moderator, the actual thermal irradiation flux across the sample might be reduced from this estimate.

Preliminary NAA experiments were performed for comparison to the calculated flux data. The thermal neutron flux, measured by dilute manganese/aluminum and gold/aluminum foils, was found to be $1.83 \times 10^8$ cm$^{-2}$ s$^{-1}$. Using a method of simultaneous equations, the fast neutron flux was calculated to be two orders of magnitude lower than the thermal. This relative thermalization is greater than expected based on the modeling efforts and requires further analysis. In order to further characterize the flux, foils of iron, bismuth, cobalt, and nickel were also irradiated. Although the absolute quantities of radionuclides present in these materials can only be approximated, the spectra are consistent with the flux data. During the half-hour irradiations, each rabbit was rotated 180 degrees at the fifteen minute mark to normalize any anomalies in the neutron field. Neutron self shielding calculations have been performed on the foil materials and agree well with the measured values. This set of simple irradiations demonstrates the analytical usefulness of such an irradiation station. Many short-lived and medium-lived nuclides are readily accessible without forming significant radioactivity in the samples. Sodium, barium, manganese, chlorine, bromine, arsenic, and antimony are particularly sensitive. Geological materials also exhibit promise and were irradiated for one hour under the conditions mentioned above. In these materials, potassium, sodium, manganese, and some rare earths such as lanthanum were identified in the spectra.

## 4.3 PRELIMINARY PGNAA EXPERIMENTS

First tests with the system shown in Figure 3 indicated the detector count rate and dead time were too high with any sizeable $^{252}$Cf source, therefore more lead shielding was added around the detector. Although PGNAA spectra were obtained for the primary system components (hydrogen and lead), adequate sensitivity for practical sample analysis was never achieved because of equipment limitations. Fulfillment of current plans for equipment upgrades is expected to provide a practical in-cell PGNAA system for research and chemical analysis.

# 5. CONCLUSIONS

The variety of irradition configurations available within the CUF provides a useful suite of choices to meet sample irradiation needs. Reasonable agreement was obtained between calculations, experiments, and earlier publications with respect to the thermal neutron flux achievable from high-intensity Cf sources. More detailed analysis is required to correlate computational and experimental results for the fast neutron flux. Assuming a 50-mg limit on

total $^{252}Cf$, the polyethylene moderator configurations maximize the thermal neutron flux better than the pool for source-to-sample distances of 7 cm or less; this is consistent with Nichols' earlier calculations.[10]

Future incorporation of gamma spectroscopy capabilities within the CUF to provide INAA and PGNAA complements to the existing NAA capabilities will greatly enhance the experimental versatility of the CUF. The analyses of in-pool NAA capabilities presented here will provide useful insight into optimizing the irradiator design for the future FDA INAA facility at the CUF. The capability currently exists to "brute force" the existing storage pool configuration in the CUF to match or exceed the optimized thermal flux of $2 \times 10^9$ $cm^{-2}$ $s^{-1}$ in the SRL INAA facility by using several hundred milligrams of $^{252}Cf$ campaign material. Selection of an optimized irradiator design for the future INAA facility will provide a premier $^{252}Cf$-based facility with neutrons always available on demand.

## REFERENCES


1. J. B. Knauer and R. C. Martin, "Californium-252 Production and Neutron Source Fabrication," in *Californium-252. Isotope for 21st Century Radiotherapy*, J. G. Wierzbicki, ed., Kluwer Academic Publishers, 1997, pp. 7-24.

2. I. W. Osborne-Lee and C. W. Alexander, "CALIFORNIUM-252: A Remarkable Versatile Radioisotope," Oak Ridge National Laboratory Report ORNL/TM-12706, 1995.

3. K. W. MacMurdo and W. W. Bowman, "Automated Absolute Activation Analysis with Californium-252 Sources," Savannah River Laboratory Report DP-1457, 1978.

4. R. A. Sigg, "Neutron Activation Analysis with $^{252}Cf$ Neutron Source at Savannah River Site," *Trans. Am. Nucl. Soc.* **68(A)** (1993) 140.

5. G. Gleason, "INAA on Environmental Samples at Oak Ridge Associated Universities," Abstract from the *Californium-252 Workshop*, Oak Ridge National Laboratory, Oak Ridge, Tenn., April 13-14, 1988.

6. P. Shea, T. Gozani, and H. Bozorgmanesh, "A TNA explosives-detection system in airline baggage," *Nucl. Instr. and Meth. A* **299** (1990) 444.

7. EG&G ORTEC, *PINS – Portable Isotopic Neutron-Spectroscopy Chemical Assay System*, sales brochure, 1996.

8. S. Reucroft, R. Rusack, D. Ruuska, and J. Swain, "Neutron irradiation damage of APD's using $^{252}Cf$," *Nucl. Instr. and Meth. A* **394** (1997) 199.

9. T. E. Byrne, L. F. Miller, R. Martin, D. Duckworth, M. Terzaghi-Howe, and G. W. Kabalka, "Determination of Uptake and Efficacy of Compounds for Boron Neutron Capture Therapy," presented at the Health Physics Society Annual Meeting, San Antonio, Texas, June 29 - July 3, 1997.

10. J. P. Nichols, "Design Data for $^{252}Cf$ Neutron Source Experiments," *Nucl. App.* **4** (1968) 382.

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