

RECEIVED
JUL 08 1996
OSTI

Measurement of ^{235}U Content and Flow of UF_6 Using Delayed Neutrons or Gamma Rays Following Induced Fission

D. C. Stromswold
A. J. Peurrung
P. L. Reeder
R. W. Perkins

June 1996

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle



MASTER MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831;
prices available from (615) 576-8401.

Available to the public from the National Technical Information Service,
U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161

This document was printed on recycled paper.

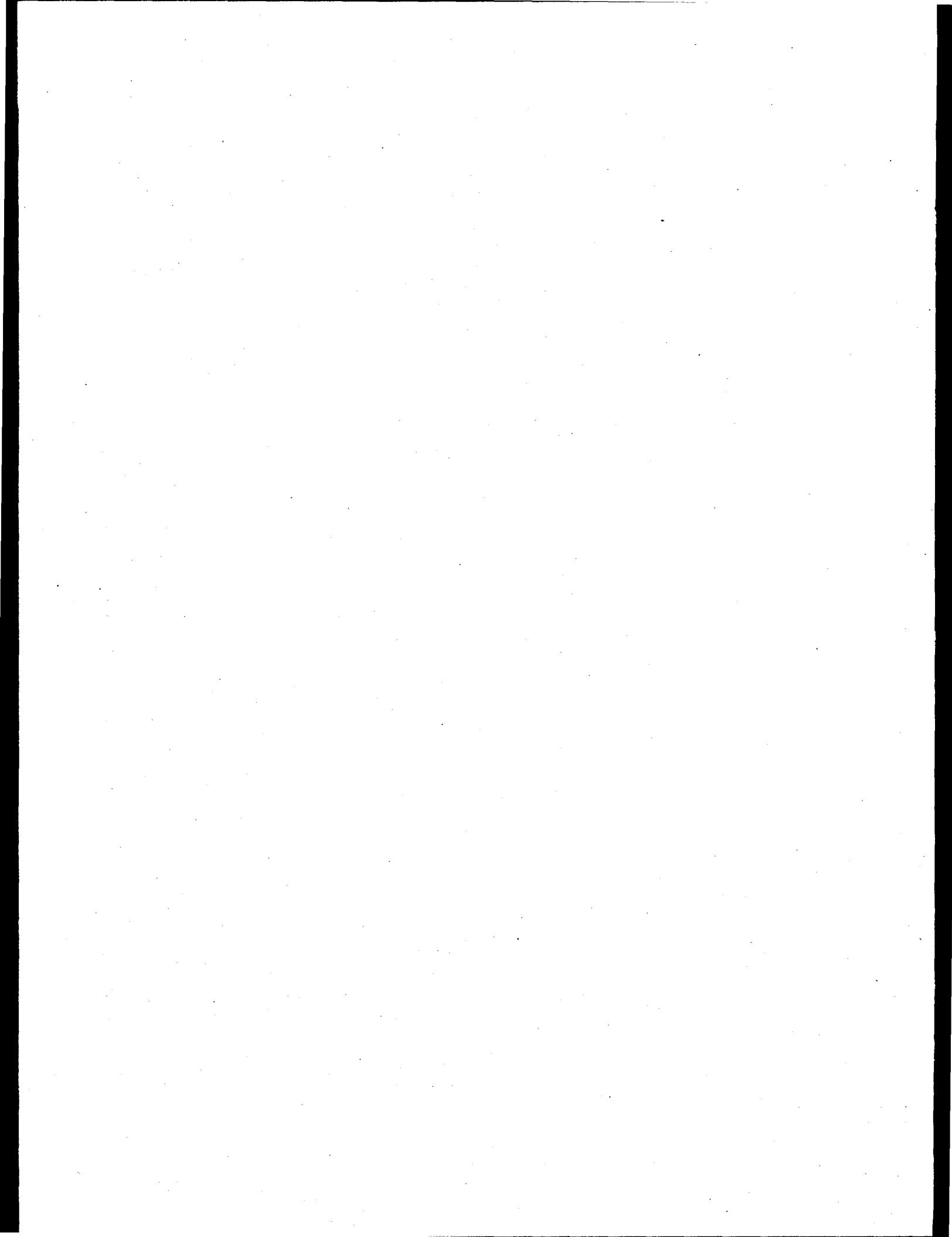
Measurement of ^{235}U Content and Flow of UF_6 Using Delayed Neutrons or Gamma Rays Following Induced Fission

D. C. Stromswold
A. J. Peurrung
P. L. Reeder
R. W. Perkins

June 1996

Prepared for
the U. S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest National Laboratory
Richland, Washington 99352



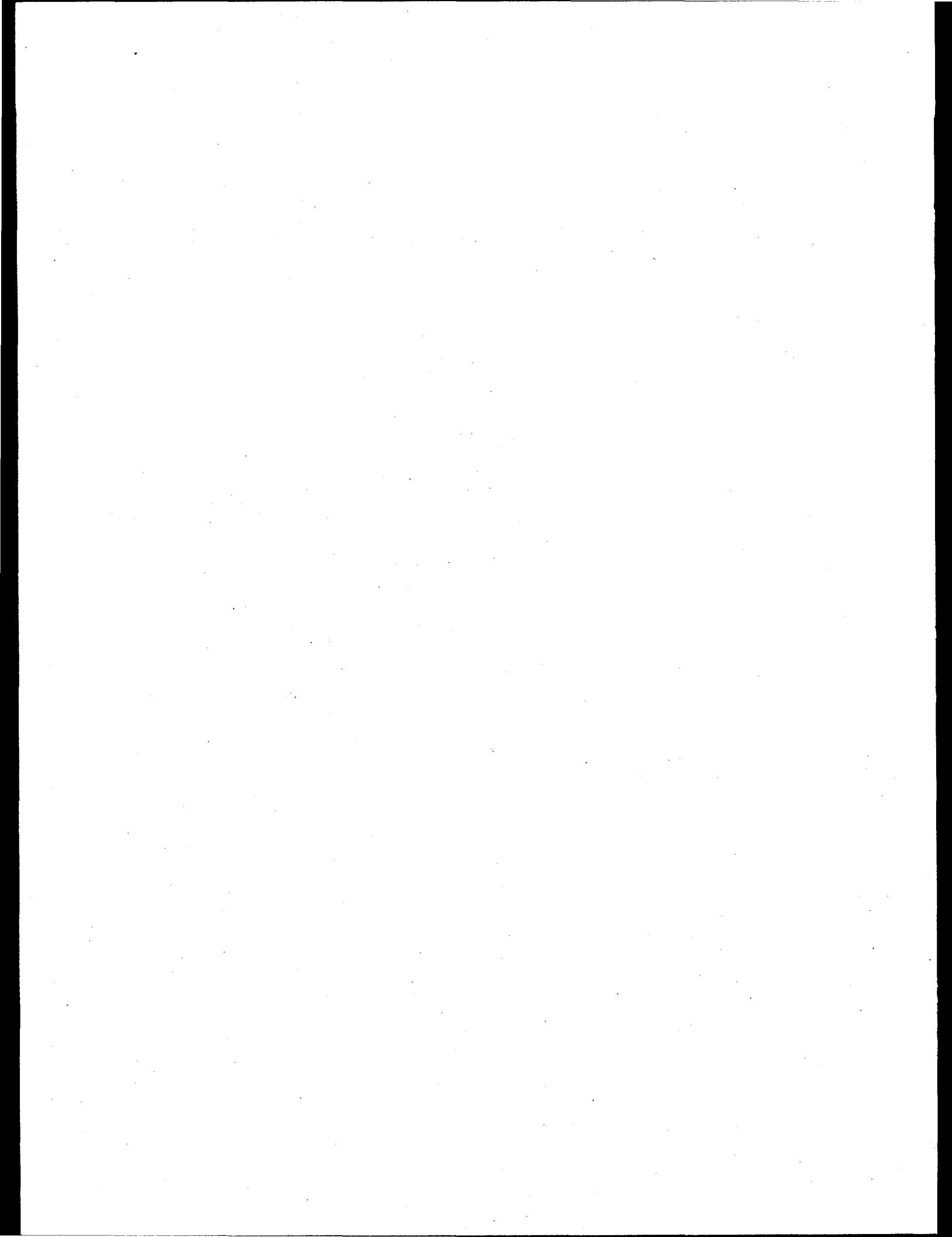
Summary

Feasibility experiments conducted at Pacific Northwest National Laboratory demonstrate that either delayed neutrons or energetic gamma rays from short-lived fission products can be used to monitor the blending of UF₆ gas streams. A ²⁵²Cf neutron source was used to induce ²³⁵U fission in a sample, and delayed neutrons and gamma rays were measured after the sample moved "downstream." The experiments used a UO₂ powder that was transported down the pipe to simulate the flowing UF₆ gas. Computer modeling and analytic calculation extended the test results to a flowing UF₆ gas system.

Neutron or gamma-ray measurements made at two downstream positions can be used to indicate both the ²³⁵U content and UF₆ flow rate. Both the neutron and gamma-ray techniques have the benefits of simplicity and long-term reliability, combined with adequate sensitivity for low-intrusion monitoring of the blending process. Alternatively, measuring the neutron emission rate from (a, n) reactions in the UF₆ provides an approximate measure of the ²³⁵U content without using a neutron source to induce fission.

The implementation for blend monitoring will place a ²⁵²Cf neutron source and moderator (polyethylene) adjacent to the pipes carrying the UF₆ gas. Neutron detectors located downstream from the neutron source will measure the delayed neutrons emitted as the result of the induced fission. Measurements made at two different downstream locations will be used to establish the flow rate as well as measure ²³⁵U content. Additional detectors located upstream from the neutron source will provide a measure of the neutron background for subtraction during data analysis. At a minimum, the installation will include monitoring of the pipes carrying the highly enriched gas to the blending point.

Based on the successful completion of the feasibility study, it is recommended that testing with actual UF₆ gas flow be conducted at the Portsmouth Gaseous Diffusion Plant.



Contents

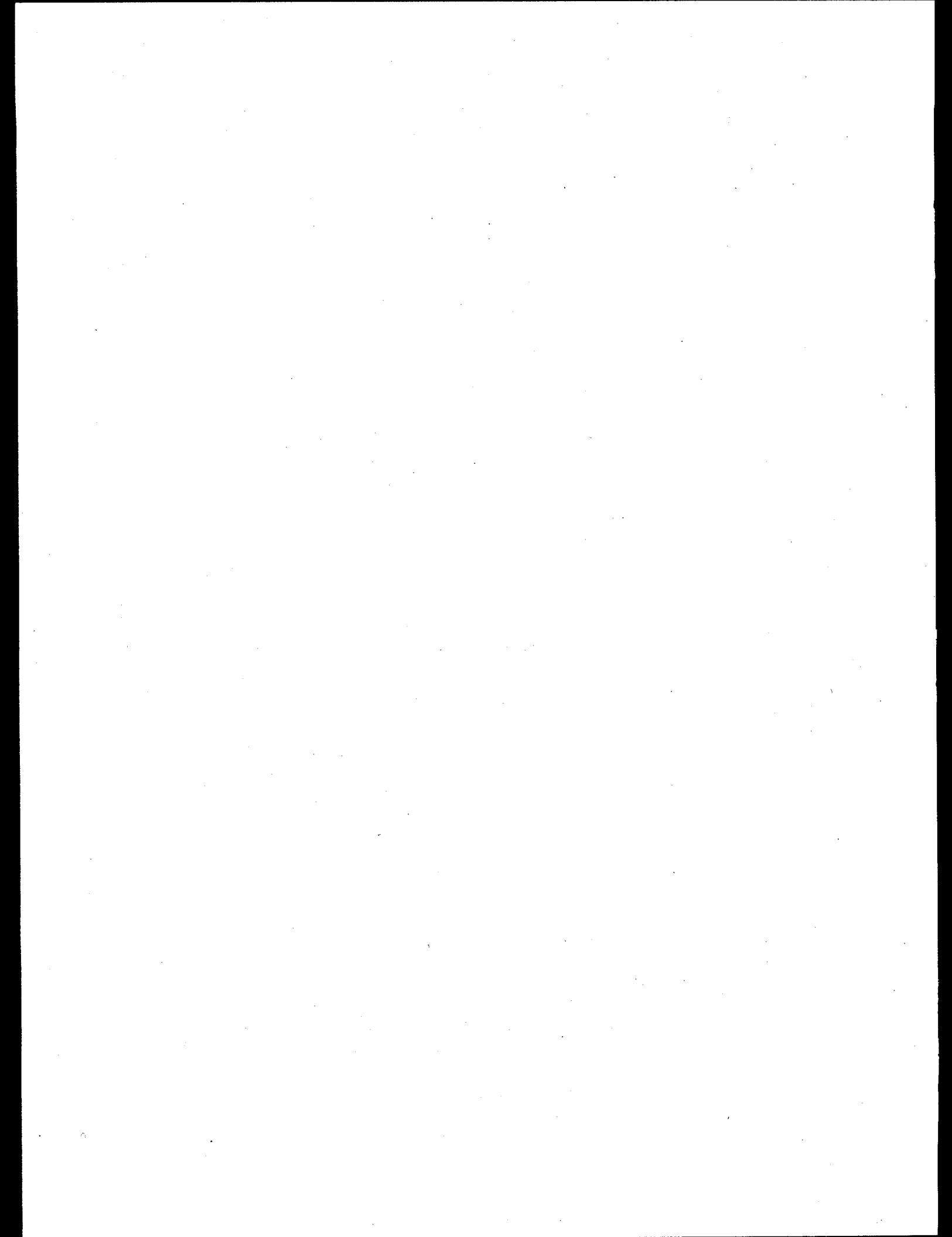
Summary	iii
Introduction	1
Feasibility Experiments	5
Equipment and Setup	5
Procedures and Results	6
Computer Simulation of UF ₆ Gas System	11
Simulation Geometry	11
Induced Fissions and Background	12
Flow Speed	15
Conceptual Deployment	17
Conclusions	19
References	19

Figures

1. Blending of UF ₆ Gas Streams Having Low and High Enrichment of ²³⁵ U	1
2. Fission of ²³⁵ U Produces Prompt Neutrons and Fission Products	2
3. Experimental Arrangement	5
4. Electronics for ³ He Neutron Detectors and NaI Gamma-Ray Detector	6
5. Neutron Counts Following Induced Fission of ²³⁵ U	7
6. Gamma-Ray Counts Following Induced Fission of ²³⁵ U	9
7. MCNP Geometry for Blending Simulation	12
8. Dependence of Count Ratio on Flow Speed	16
9. Conceptual Arrangement of Neutron Detectors on Blending Line	17

Tables

1. UF ₆ Flow Parameters for Anticipated Blending	1
2. Six-Group Representation of Delayed Neutrons from Fissioning of ²³⁵ U	2
3. Neutron-Counting Results from Cyclic Irradiation of UO ₂ Source	8
4. Neutron Count Rates Without ²⁵² Cf Irradiation Source	8
5. Gamma-Ray Counting Results from Cyclic Irradiation of UO ₂ Source	9
6. Comparison of Feasibility Test Parameters and Anticipated Blending Conditions	11
7. MCNP Computer Simulation Results	13
8. Fraction of Delayed Neutrons (DN _d) Emitted At Various Source-to-Detector Distances and Flow Velocities	14



Introduction

Because highly enriched uranium can present control and non-proliferation difficulties, the ^{235}U content of some enriched uranium is being reduced to decrease the inventory, but leave enough enrichment for power production. A product stream with the desired enrichment can be produced by blending two gas streams of UF_6 , one containing highly enriched uranium and the other low enrichment. Research in monitoring the blending of high- and low-enrichment uranium was conducted at Pacific Northwest National Laboratory^(a) for the U.S. Department of Energy, Office of Research and Development (NN-20). Figure 1 shows the blending of two gas streams of UF_6 , one containing highly enriched uranium and the other containing low enrichment. The product stream of the mixed gas contains uranium with enrichment of about 4.4 %. Table 1 gives flow parameters for the input and output streams. Monitoring the ^{235}U content of the input stream having the highly enriched uranium will provide confidence that high-enrichment uranium is being used in the blending process, and monitoring the output stream will provide an on-line measure of the ^{235}U in the mixed product.

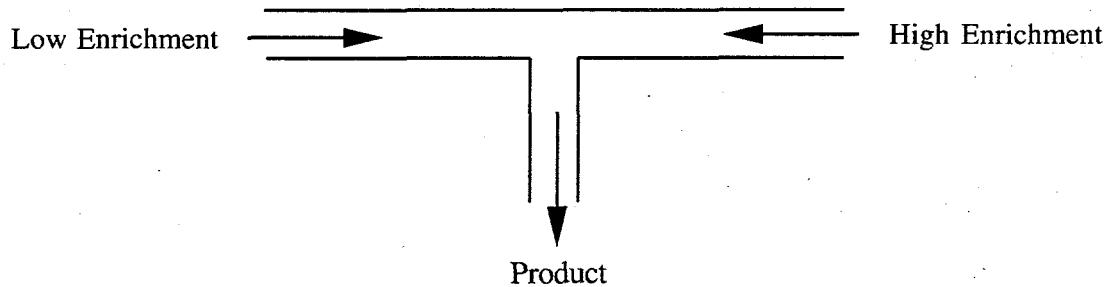


Figure 1. Blending of UF_6 Gas Streams Having Low and High Enrichment of ^{235}U

Table 1. UF_6 Flow Parameters for Anticipated Blending

Stream	Speed (m/s)	Mass Flow (kg/s)	Pressure (mm Hg)	Enrichment (%)
Input (high ^{235}U)	0.06	0.0007	38	90
Input (low ^{235}U)	1.4	0.02	45	1.5
Output (product)	1.02	0.021	30	4.4

Inducing fission in the ^{235}U gives a method for monitoring the ^{235}U content and the flow speed of the UF_6 gas. An external, moderated ^{252}Cf source provides thermal neutrons that interact with ^{235}U , inducing fission. Although neutrons are emitted promptly during the fission, some additional neutrons are emitted by the fission products at a delayed time (Figure 2). The time delay allows the fission products (and the UF_6 gas) to move downstream to the location of a detector. Table 2 shows the six major groupings of delayed neutrons emitted from the fissioning of ^{235}U . If two detectors are located at different downstream distances, the ^{235}U content and flow speed can be estimated from the separation distance of the detectors, the known half-lives of the fission products, and the observed

(a) Pacific Northwest National Laboratory is operated for the U.S. Department of Energy by Battelle under Contract DE-AC06-76RLO 1830.

count rate at the first and second detectors. Containers filled with ${}^3\text{He}$, providing long-term, reliable operation, can be used to monitor the delayed neutrons.

The gamma rays emitted by the highly radioactive fission products also provide a means for measuring ${}^{235}\text{U}$ content and flow speed following induced fission. Locating gamma-ray detectors downstream allows measurement of the gamma-ray activity in a manner similar to that of the delayed neutrons. By counting only the high-energy gamma rays emitted following fission, potential signal interference from plateout of fission products on the inner walls of the pipes can be avoided. The gamma-ray detectors can be simple scintillation detectors that do not require cooling, and they can operate in the gross counting mode with a high discrimination level to reject low-energy gamma rays. These detectors are reliable for long-term operation and require no maintenance.

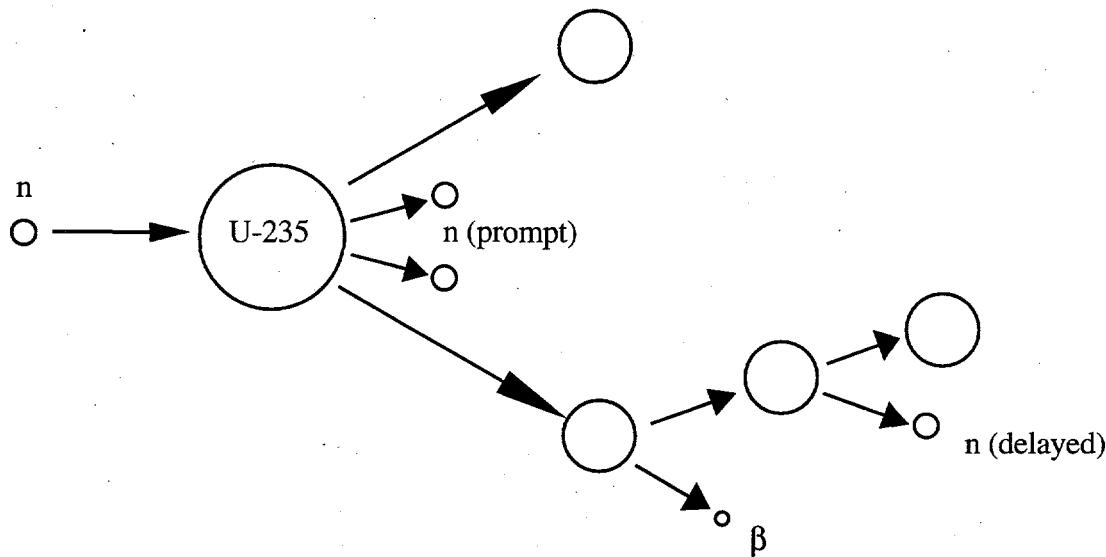


Figure 2. Fission of ${}^{235}\text{U}$ Produces Prompt Neutrons and Fission Products, Some of Which Can Subsequently Produce Delayed Neutrons

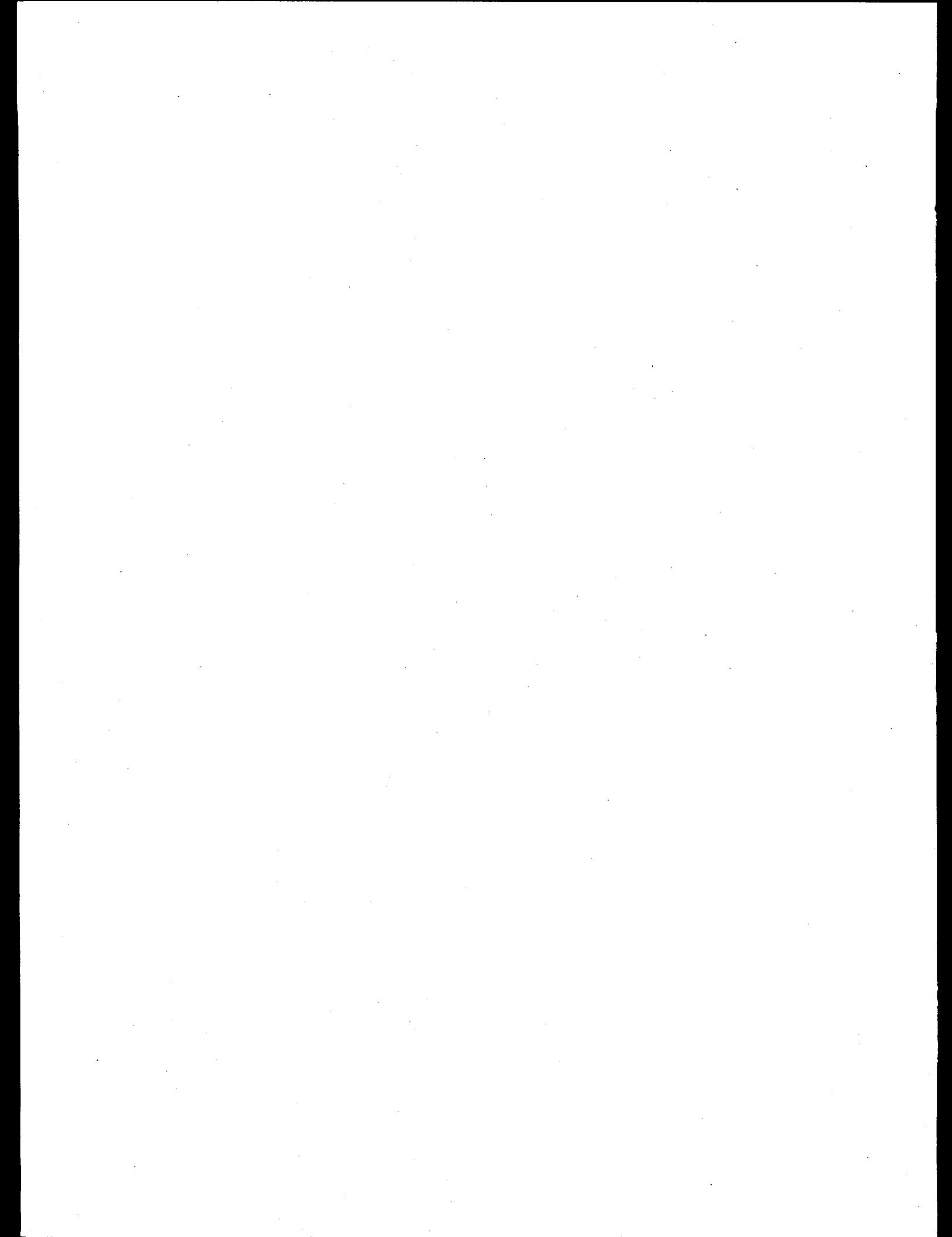
Table 2. Six-Group Representation of Delayed Neutrons from Fissioning of ${}^{235}\text{U}$

Group	Half-Life(s)	Relative Abundance
1	52.12	0.0380
2	21.33	0.1918
3	5.686	0.1638
4	2.187	0.3431
5	0.7011	0.1744
6	0.2346	0.0890

A third method for monitoring ${}^{235}\text{U}$ content detects neutrons emitted from (α, n) reactions in the UF_6 gas. Rates of neutron emission differ between ${}^{235}\text{UF}_6$ and ${}^{238}\text{UF}_6$, allowing them to be

distinguished. Monitoring (a,n) reactions requires no neutron source to induce fission, but no measure of flow speed is provided.

Laboratory tests with detectors and samples containing ^{235}U show the feasibility of the proposed methods for monitoring the UF_6 blending process. In these tests, a sample of enriched uranium was moved to simulate gas flow. Irradiating the sample with neutrons produced fissions, and then the sample moved through a pipe to locations adjacent to neutron and gamma-ray detectors. Analytical calculations and computer simulations with the code MCNP extended the test results to conditions applicable to flowing UF_6 gas and anticipated blending conditions.



Feasibility Experiments

Equipment and Setup

A UO_2 powder sample is used in the feasibility tests to simulate the UF_6 gas. The sample has 86 g of uranium at an enrichment of 2.4 %. The sample moves inside a square aluminum pipe with a wall thickness of 3.2 mm and an inside dimension of 5.7 x 5.7 cm. The source moves 3 m for the delayed neutron measurements and 2.3 m for the gamma-ray measurements.

Figure 3 shows the experimental arrangement of the aluminum pipe, UO_2 sample, neutron and gamma-ray detectors, and the irradiating ^{252}Cf neutron source. The neutron detector array (Reeder et al. 1977) consists of 42 ^3He tubes (4 atm., 2.5 cm diameter, 36 cm length) embedded in polyethylene around the pipe. The individual tubes are parallel to the pipe and arranged in concentric circles in holes in the polyethylene. The detection efficiency for delayed fission neutrons is about 45%, based on prior experiments. The tubes are electrically connected in parallel, in groups of 12 tubes, except for one group that has 6 tubes. Figure 4 shows the electronics used to process the signals from the detectors. High discriminator settings on the single channel analyzer (SCA) output signals ensure that gamma-rays are not also being counted by the ^3He detectors. The Fan In/Fan Out unit shown in Figure 4 produces a logic output pulse whenever any one of the four input neutron signals occur. The scaler sums the neutron counts until it is polled by the computer.

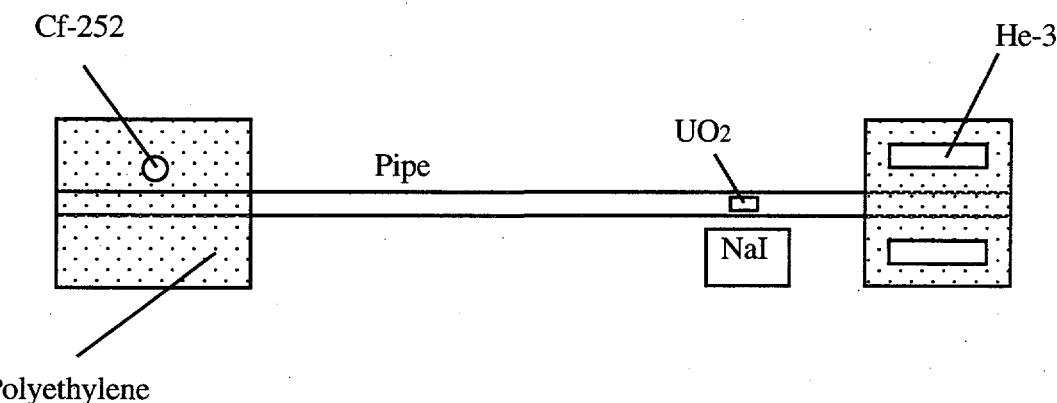


Figure 3. Experimental Arrangement with ^3He Neutron Detectors, UO_2 Sample in Pipe, NaI Gamma-Ray Detector, and ^{252}Cf Neutron Source for Irradiation

The computer, an Apple Power Book 170 operating KMax acquisition software, polls the scaler every second to acquire multichannel scaling data with 1-s bin widths and stores the data in a 100-channel time-sorted spectrum. The time reference for repeated data collections is the movement of the UO_2 source through the pipe, cycling between the irradiation and counting locations.

The gamma-ray detector is a 12.7 x 5 cm (diameter x length) NaI scintillation detector shielded by 5 cm of lead to reduce background. The discriminator on the timing single-channel analyzer (TSCA) is set to either 1.4 MeV or about 3 MeV to check the signal dependence upon gamma-ray energy. A separate section of the same scaler used to collect the neutron count collects the gamma-ray counts. Gamma-ray counts are passed to the computer when the scaler is polled, and this occurs on the same time basis used for collecting the neutron counts (i.e., every second for 100 s).

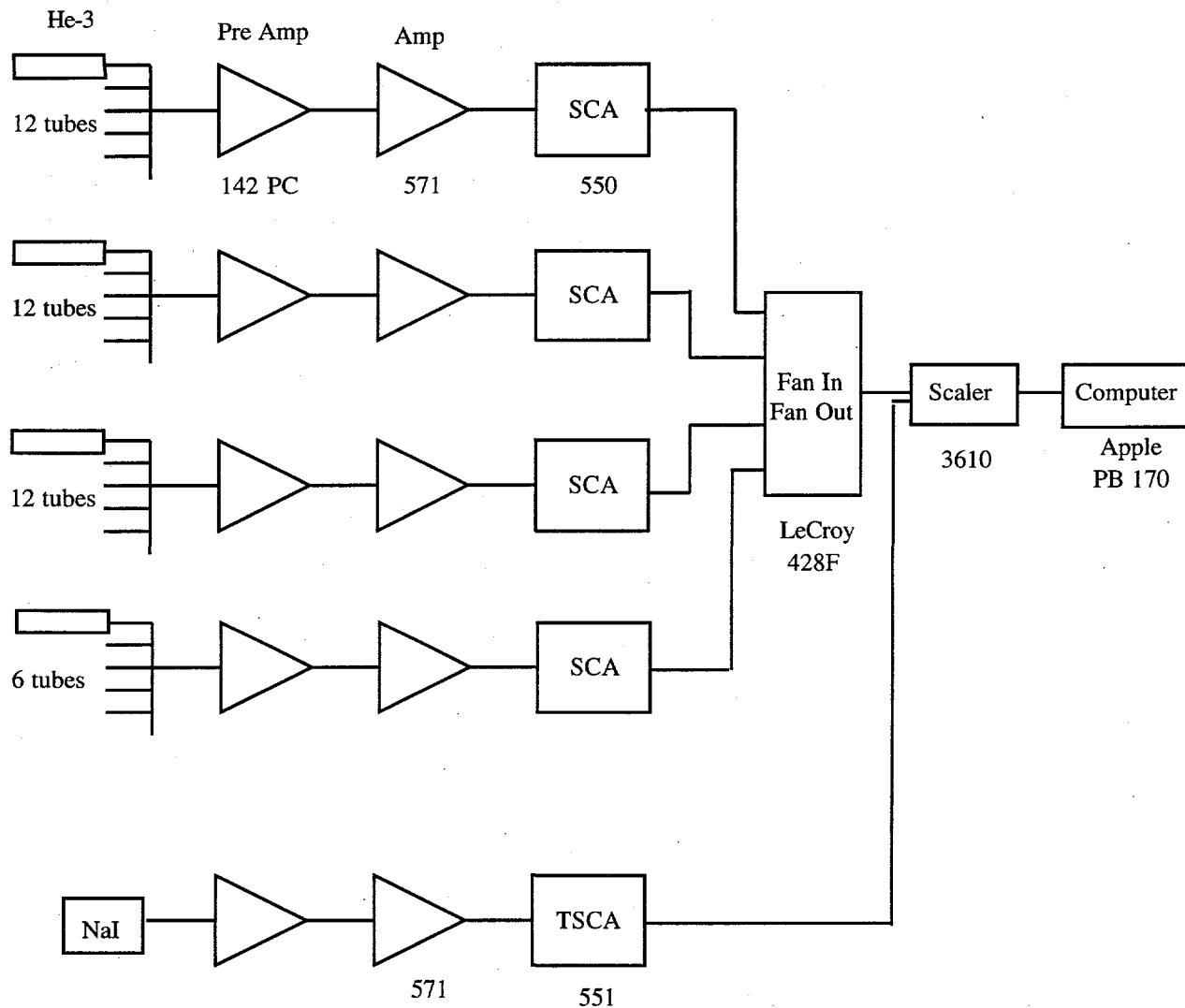


Figure 4. Experimental Arrangement with ^3He Neutron Detectors, UO_2 Sample in Pipe, NaI Gamma-Ray Detector, and ^{252}Cf Neutron Source for Irradiation

A ^{252}Cf source emitting 10^6 neutrons per second is located about 5 cm from the pipe in a polyethylene enclosure that is about 20 cm thick. The polyethylene moderates the energy of the neutrons down to thermal energy, where the neutrons can induce fission in ^{235}U . Boron-loaded paraffin (about 20 cm thick) on the outside of the polyethylene moderator, between the ^{252}Cf source and the neutron detectors, reduces the neutron background from the ^{252}Cf that would be present during the counting of delayed neutrons.

Procedures and Results

The UO_2 sample is manually moved between the counting and irradiating positions using a rod attached to the sample container. A single cycle lasts about 100 s and consists of three main time segments: counting before irradiation (from about 1 to 20 s), irradiation (from about 22 to 42 s), and counting after irradiation (from about 44 to 100 s). Transit time between the neutron counting

and irradiating positions requires about 2 to 3 s, which corresponds to a speed of about 1 to 1.5 m/s. When the counting and irradiation cycle is repeated many times in sequence to reduce statistical uncertainties, the initial counting segment (1 to 20 s) becomes essentially a wrap-around of the third counting segment, except for a delay of a few seconds at the start of each new cycle. The computer controls the data acquisition sequence and accumulates the data for repeated cycles.

Figure 5 shows the neutron time spectrum acquired by cycling the UO_2 sample between the neutron counter array and the irradiation position. The data in the figure are the sum of two separate runs, each with 10 cycles. The clear decay curve of the fission products is readily apparent in Figure 5, beginning at about 46 s, after the irradiated sample returned to the counting position. Table 3 contains the integrated neutron counts and count rates from Figure 5 over selected time intervals. The count rates are clearly distinguishable above the "background" counts, which are obtained when the sample is located at the irradiation position rather than at the counting position.

A separate measurement with both the ^{252}Cf source and the sample removed from the experimental area yields a background of about 0.8 n/s. Possible origins for this neutron background include neutron sources stored in the building and cosmic-ray interaction with heavy materials, producing neutron spallation. To check the neutron spallation rate, 11.9 kg of lead are placed in the pipe at the neutron counting location. The measured count rate (with discriminators set at about 2 V) is about 0.27 n/s above background. Assuming the neutron spallation rate is the same for lead and uranium, the contribution of cosmic-ray-induced neutron spallation from the UO_2 sample (86 g) to any background would be insignificant (< 0.002 n/s).

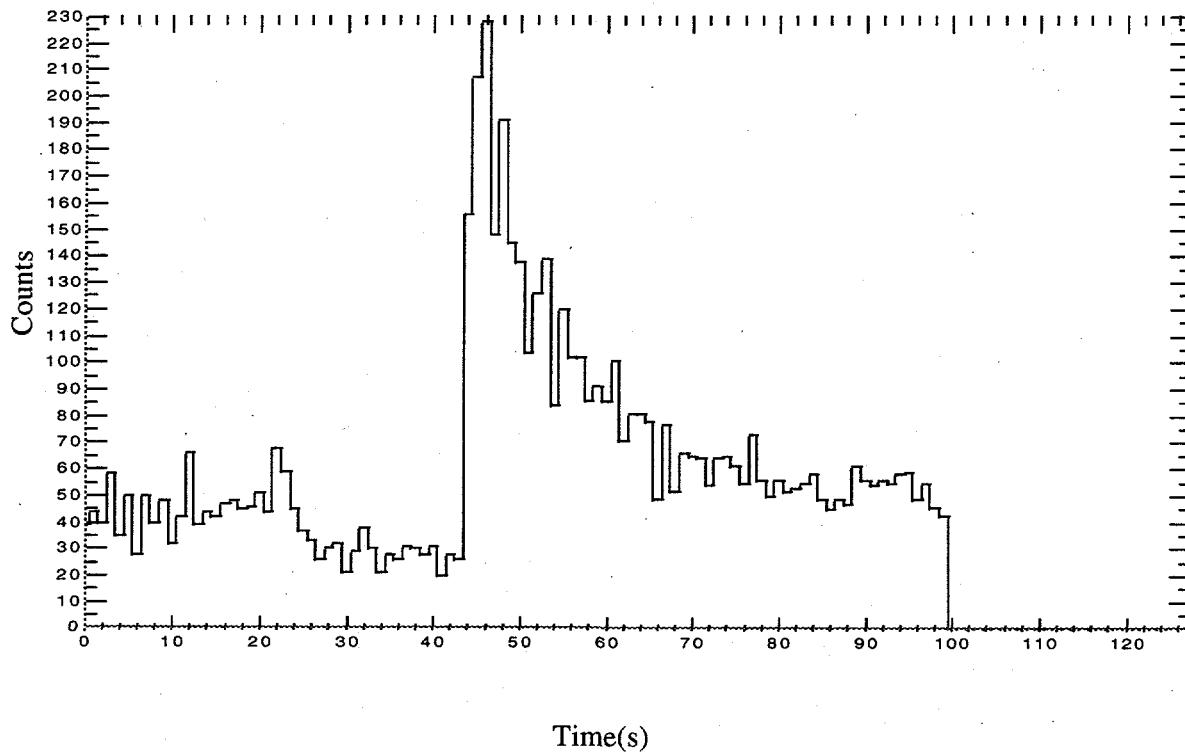


Figure 5. Neutron Counts Following Induced Fission of ^{235}U . The sample was irradiated from 22 to 42 s, and delayed neutrons were counted from 46 to 100 s.

Table 3. Neutron Counting Results from Cyclic Irradiation of UO₂ Source

Time Interval (s)	Counts	Count Rate (c/s)
44 to 54	1666	8.3
55 to 75	1616	4.0
76 to 99	1290	2.8
1 to 20 (wrap around)	895	2.4
26 to 40 (background)	434	1.6

Table 4 contains additional neutron data obtained without the ²⁵²Cf irradiation source and for various neutron discriminator settings. In addition to the results from the UO₂ sample, the table contains results from a UF₄ sample placed in the pipe near the neutron detectors. The UF₄ sample was a powder containing 150.5 g of uranium with natural enrichment. The UF₄ sample provided more counts above background than did the UO₂ sample, in accordance with expectations. The data in Table 4 are consistent with the detection of neutrons, rather than gamma-rays, by the ³He tubes. For UF₆ (not in Table 4), emitted neutrons arise mainly from (α, n) reactions, although spontaneous fission from ²³⁸U does contribute some neutrons. Including spontaneous fission, about twice as many neutrons (0.08 n/s-g) are emitted from ²³⁵UF₆ as from ²³⁸UF₆. Thus some differentiation between the two uranium isotopes is theoretically possible without inducing fission.

Table 4. Neutron Count Rates Without ²⁵²Cf Irradiation Source

Sample	Counts Per Second at Discriminator Knob Setting (V)			
	1.5	2.5	3.5	4.5
None (6kg)	1.06 \pm 0.04	1.04 \pm 0.04	0.90 \pm 0.04	0.78 \pm 0.04
UO ₂	1.69 \pm 0.08	1.45 \pm 0.07	1.31 \pm 0.07	1.16 \pm 0.06
UF ₄	3.93 \pm 0.11	3.87 \pm 0.11	3.41 \pm 0.11	2.94 \pm 0.10

Irradiating the UO₂ sample and moving it through the pipe to a location adjacent to the NaI detector produces the time spectrum of gamma rays shown in Figure 6. The data in Figure 6 are for five cycles of irradiation, using the same 100-s timing as for the neutron measurements. As with the neutron time-dependent data, a clear decay of the fission products is evident. Table 5 contains count rates for selected time intervals of the gamma-ray data in Figure 6. The discriminator setting is about 3 MeV for the gamma-ray data to avoid the lower-energy gamma rays emitted from long-lived fission products that might plateout on the pipe walls of a UF₆ flowing gas.

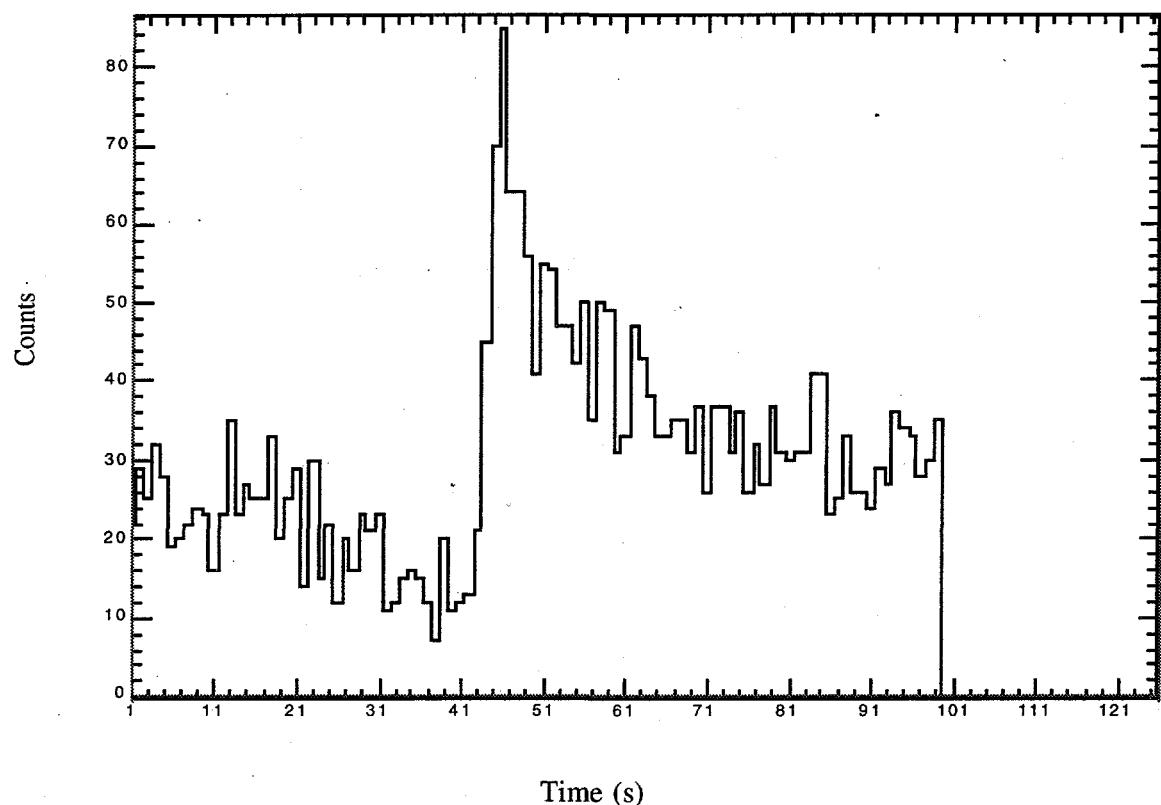
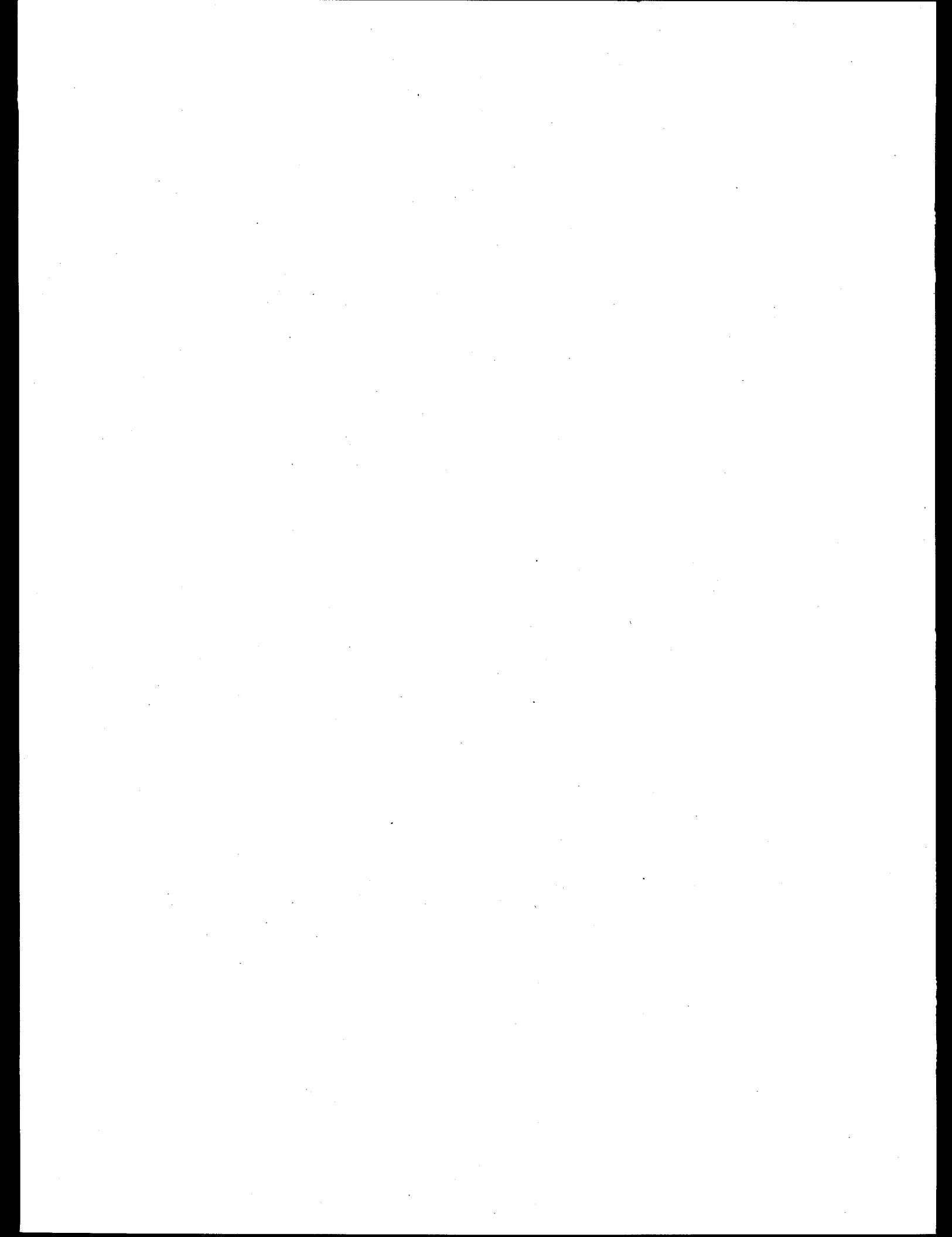


Figure 6. Gamma-Ray Counts Following Induced Fission of ^{235}U . The sample was irradiated during 22 to 42 s, and gamma rays were counted during 43 to 100 s.

Table 5. Gamma-Ray Counting Results from Cyclic Irradiation of UO_2 Source

Time Interval (s)	Counts	Count Rate (c/s)
44 to 54	628	12.6
55 to 75	789	7.9
76 to 99	736	6.4
1 to 20 (wrap around)	496	5.2
26 to 40 (background)	234	3.3



Computer Simulation of UF₆ Gas System

The experimental parameters and conditions during the laboratory feasibility tests were necessarily different from those anticipated during actual blending. The most obvious difference between the feasibility tests and blending is the use of a powder source (UO₂) rather than UF₆ as a flowing gas. Other differences include flow-pipe construction, irradiation source strength, and irradiation and counting times. Table 6 lists some of the differences between the feasibility test conditions and anticipated blending conditions.

Table 6. Comparison of Feasibility Test Parameters and Anticipated Blending Conditions

Parameter	Feasibility test	Blending
Pipe	Al with 3.2 mm wall thickness	Al or Fe with 3 to 5 mm wall
Irradiation Source	^{252}Cf emitting 10^6 n/s	^{252}Cf or other source emitting up to 5×10^7 n/s
Distance Between Irradiation Source and Neutron Detector	3 m	3 m
Irradiation and Counting Mode	pulsed	continuous
Speed During Movement from Irradiation to Counting	1.5 m/s	0.06 to 1.5 m/s
Speed During Irradiation and Counting	0	0.06 to 1.5 m/s
Neutron Detector Efficiency	~45 %	~45 %
Gamma-Ray Detector Efficiency	~5 %	~20 %
Neutron Background	~1 n/s	~1 n/s
^{235}U in Sample Volume	2 g (in UO ₂ powder)	3 g (in UF ₆ high enrich input)

Simulation Geometry

The induced fission rate in the UF₆ gas system can be estimated by computer modeling with the code MCNP (Briesmeister 1993). Figure 7 shows the geometry of the modeled system, including pipe, UF₆ gas, irradiation source, neutron moderator and shielding, and detector region. In the modeling, UF₆ gas is included as low density material (gas at 100 mm Hg and 300° K) filling an iron pipe of wall thickness 0.6 cm and outside diameter 10 cm. The iron in the pipe absorbs neutrons, reducing the number of thermal neutrons available to induce fission in the UF₆ gas. The 0.6-cm wall thickness used in the MCNP calculations is thicker than that expected to be encountered and thus produces a conservative value for the induced fission rate.

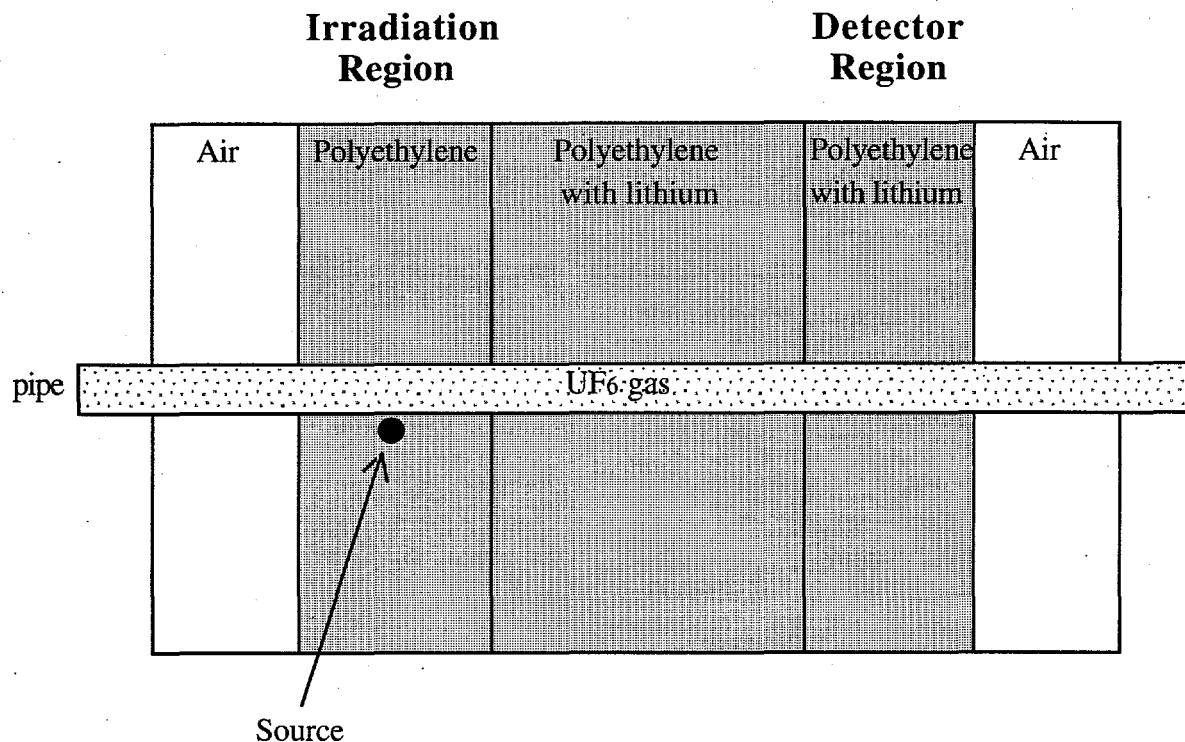


Figure 7. MCNP Geometry for Blending Simulation

The irradiation zone in Figure 7 is modeled as a 60-cm-long section of the pipe surrounded by polyethylene of radius 1 m containing a neutron source located 5 cm from the pipe and centered along the length of the polyethylene. Other radial distances of the source from the pipe are also modeled, including locating the source immediately adjacent to the pipe. The 5-cm distance provides the best thermal neutron flux for inducing fission while reducing fast neutron transmission down the inside of the pipe to the detector region, a major source of background neutrons. The 5-cm set-back distance in the polyethylene also reduces the number of high-energy neutrons that can reach the gas and potentially induce fission in ^{238}U . The modeled neutron source is a fission spectrum, which approximates a ^{252}Cf source. Other zones included in the model are filled with polyethylene containing ^{6}Li (1 atom percent) for neutron absorption. These zones provide neutron shielding between the source and detector. However, the middle zone containing polyethylene/ ^{6}Li adds little shielding, as replacing that material with air shows in one calculation.

Induced Fissions and Background

The MCNP calculations provide the number of induced fissions (per source neutron) in UF_6 gas in the irradiation section near the neutron source, assuming all uranium is ^{235}U . The calculations also give the number of neutrons from the source that reach the detector region and contribute to the background. Most of these background neutrons migrate down the pipe, rather than through the polyethylene moderator. Table 7 contains MCNP results for selected source-to-detector spacings and special conditions. As shown in the table, the induced fissions in the $^{235}\text{UF}_6$ are about 0.002 per neutron from the ^{252}Cf source. The migration of source neutrons down the pipe to the detector region decreases for increasing source-to-detector distances. Also shown in the table are the induced fissions in $^{238}\text{UF}_6$, and they are much smaller than the corresponding number of fissions in $^{235}\text{UF}_6$, as expected.

Table 7. MCNP Computer Simulation Results

Gas	Source-to-Detector Distance (m)	Induced Fissions (per 100 source neutrons)	Background Neutrons at Detector (per 100 source neutrons)
$^{235}\text{UF}_6$ @ 100 mm Hg	1	0.21	0.009 ± 0.0009
$^{235}\text{UF}_6$ @ 100 mm Hg	1.5	0.21	0.0009 ± 0.0002
$^{235}\text{UF}_6$ @ 100 mm Hg	2	0.21	0.0003 ± 0.0001
$^{235}\text{UF}_6$ @ 100 mm Hg	2.5	0.21	0.00006 ± 0.00003
$^{235}\text{UF}_6$ @ 1 mm Hg	1	0.0044	0.17
$^{238}\text{UF}_6$ @ 100 mm Hg	1	0.0006	0.11

Using the MCNP-provided number of fissions induced in the UF_6 gas, the number of delayed neutrons emitted in the detector region can be estimated analytically, based on the flow speed of the gas and the known decay rate for the ^{235}U fission products. Adding an assumed detection efficiency then provides the expected counting rate in the detectors. The equation used to obtain the count rate in the detectors is

$$\text{Rate} = SO \times IF \times DN \times DN_d \times Eff \times Enr \quad (1)$$

where

SO = neutron irradiation source output (n/s)

IF = induced fissions in $^{235}\text{UF}_6$ gas (per source neutron)

DN = fraction of induced fissions that lead to delayed neutron emission (all times) = 0.0167

DN_d = fraction of delayed neutrons emitted when gas and fission products are at detector location

Eff = detector efficiency

Enr = ^{235}U enrichment (1= 100%)

The fraction of induced fissions in ^{235}U that lead to delayed neutron emission, DN , is 0.0167, based on published data for ^{235}U (Kinsey 1979). The fraction of these delayed neutrons emitted over a given time interval, DN_d , is obtained from the abundances (a_i) and half-lives (T_i) of the fission decay products (Table 2) and the time required for the gas and fission products to move by the detector location:

$$DN_d = \sum_{i=1}^6 \int_{t_1}^{t_2} a_i \exp(-0.693 t / T_i) dt \quad (2)$$

For an assumed flow speed of 1 m/s, the irradiated section of gas (and its resultant fission products) travels 3 m from the source to the detector in 3 s. Table 8 gives the values of DN_d for the time interval that the gas would be within ± 0.25 m of the center of the detector region for various source-to-detector spacings.

Table 8. Fraction of Delayed Neutrons (DN_d) Emitted at Various Source-to-Detector Distances and Flow Velocities

Speed (m/s)	Source-to-Detector Distance (m)			
	1	2	3	4
0.06	0.0625	0.0234	0.0128	0.0078
1	0.0916	0.0523	0.0355	0.0261

The detector efficiency, Eff , varies with the detector type and configuration around the pipe. An array of ^3He tubes embedded in polyethylene would have an efficiency of at least 30%. The ^{235}U enrichment, Enr , in the UF_6 gas is between 1.5 and 90% (Table 1).

Selecting, for example, an enrichment of 4.4%, and using the MCNP result that about 0.21% of the source neutrons produce fission in the ^{235}U (Table 7), the following estimated count rate results for a 3-m source-to-detector spacing using Equation 1:

$$\begin{aligned} \text{Rate} &= SO \times 0.0021 \times 0.0167 \times 0.0355 \times 0.3 \times 0.044 \\ \text{Rate} &= SO \times (1.6 \times 10^{-8}) \end{aligned}$$

Assuming $SO=10^7$ n/s and counting for 10^4 s, the following counts would be expected in the detector:

$$\begin{aligned} \text{Counts} &= \text{Rate} \times \text{Time} \\ \text{Counts} &= [10^7 \times (1.6 \times 10^{-8})] \times 10^4 \\ \text{Counts} &= 1600 \end{aligned}$$

For a gas pressure of 30 mm Hg (rather than 100 mm Hg) the counts are 490.

The background count from the source during this same counting time is

$$\begin{aligned} \text{Bkg} &= \text{Bkg Rate} \times \text{Time} \\ \text{Bkg} &= (SO \times M \times Eff) \times \text{Time} \\ \text{Bkg} &= (10^7 \times 4 \times 10^{-7} \times 0.3) \times 10^4 \\ \text{Bkg} &= 12,000 \end{aligned}$$

where M is the fraction of fast neutrons from the source that migrate through the pipe to the detector region ($M = 4 \times 10^{-7}$ at 3 m, by extrapolation of background results in Table 7).

The natural background (not due to migration down the pipe) produces an additional ~ 1 c/s, or 10,000 counts in 10^4 s, based on the feasibility experiments. Therefore, the total background counts = 22,000 in 10^4 s. Methods for monitoring this background are discussed in a following section.

The statistical uncertainty, σ , (1 standard deviation) for 600 counts and a background of 40,000 counts is

$$\sigma = \sqrt{(Cts + Bkg) + Bkg}$$
$$\sigma = \sqrt{(490 + 22000) + 22000}$$
$$\sigma = 210$$

The fractional uncertainty is then

$$\sigma/Cts = 210/490$$

$$\sigma/Cts = 43\%$$

For the high-enrichment input blending line, the flow speed is small, resulting in the loss of many delayed neutrons before the fission products reach the detector region. However, the high enrichment compensates for the low speed by providing more initial fissions in the gas. Using a flow speed of 0.06 m/s, enrichment of 90%, source-to-detector spacing of 3 m, and gas pressure of 40 mm Hg, the counts in 10^4 s are 4800, and the background is 22,000 counts (migration from the source plus expected natural background). This produces a fractional uncertainty of 5%. If the counting time is decreased to 10^3 s, the statistical uncertainty becomes 14%.

The MCNP results also show that about 10% of the source neutrons are absorbed in the walls of the pipe, decreasing the neutrons available to induce fission in the UF_6 gas. Because the pipe absorbs thermal neutrons that would otherwise be available to induce fission in the UF_6 , the effect on the induced fission rate can be significant. If the pipe is made of aluminum, rather than steel, the neutron absorption will be less, and the induced fission rate will be larger.

Another important effect is the loss of fission products to the walls of the pipe. Most fission products are non-gaseous and will stick to any surface they encounter. The range of fission fragments in UF_6 can be estimated from published tables (Northcliffe and Schilling 1970). The light-mass fragments have the greatest range and also produce almost 70% of the delayed neutrons (Reeder and Warner 1984). An estimate of the recoil range of a typical light-mass fission fragment is about 5 cm in UF_6 at the density of the high-enrichment gas stream. Clearly many of the fission products will reach the wall and be lost to the downstream flow. Detailed calculations are needed to determine the actual fraction that recoils to the walls. If the gas flow in the pipe is turbulent, additional fission products may stick to the pipe and be lost as well. These effects should be evaluated under realistic conditions.

Flow Speed

Measuring the counts at two downstream locations provides a means for calculating the flow speed of the gas containing the fission products. The neutron counts at the two downstream detectors, after background subtraction, are

$$Count_1 = Rate_1 \times time$$
$$Count_2 = Rate_2 \times time$$

Using Equation (1), and assuming the same counting time and detector efficiency for both detectors, the ratio of counts in the two detectors is

$$\begin{aligned}\text{Count}_1/\text{Count}_2 &= \text{Rate}_1/\text{Rate}_2 \\ \text{Count}_1/\text{Count}_2 &= \text{DN}_{d,1}/\text{DN}_{d,2}\end{aligned}$$

where $\text{DN}_{d,1}$ is the fraction of delayed neutrons emitted when the fission products are at the location of the first detector, and $\text{DN}_{d,2}$ is the corresponding quantity for the second detector. Evaluating $\text{DN}_{d,1}$ and $\text{DN}_{d,2}$ from Equation (2) gives

$$\frac{\text{Count}_1}{\text{Count}_2} = \frac{\sum_{i=1}^6 \int_{t_1}^{t_2} a_i \exp(-0.693 t_i^{(1)}/T_i) dt_i^{(1)}}{\sum_{i=1}^6 \int_{t_1}^{t_2} a_i \exp(-0.693 t_i^{(2)}/T_i) dt_i^{(2)}} \quad (3)$$

where

$t^{(1)}$ = time for fission products to reach location of detector 1

$t^{(2)}$ = time for fission products to reach location of detector 2.

The times, $t^{(1)}$ and $t^{(2)}$, are related to the flow speed, v , and the distances, D_1 and D_2 , between the source and the first and second detectors:

$$t^{(1)} = D_1/v$$

$$t^{(2)} = D_2/v.$$

Figure 8 shows a plot of Equation (3) for the case $D_1=3$ m and $D_2 = 6$ m. As an example of using this figure to obtain the flow speed, suppose the ratio of counts in the two detectors is 2. Entering the plot at Count ratio=2 and following across to the curve, then down to the velocity axis, shows that the flow is 0.13 m/s. Statistical uncertainties in the count ratio produce uncertainties in the calculated flow speeds, as can be seen by entering the figure at different count ratio values. Above about 0.2 m/s, the flatness of the curve can produce large uncertainties in the calculated speeds.

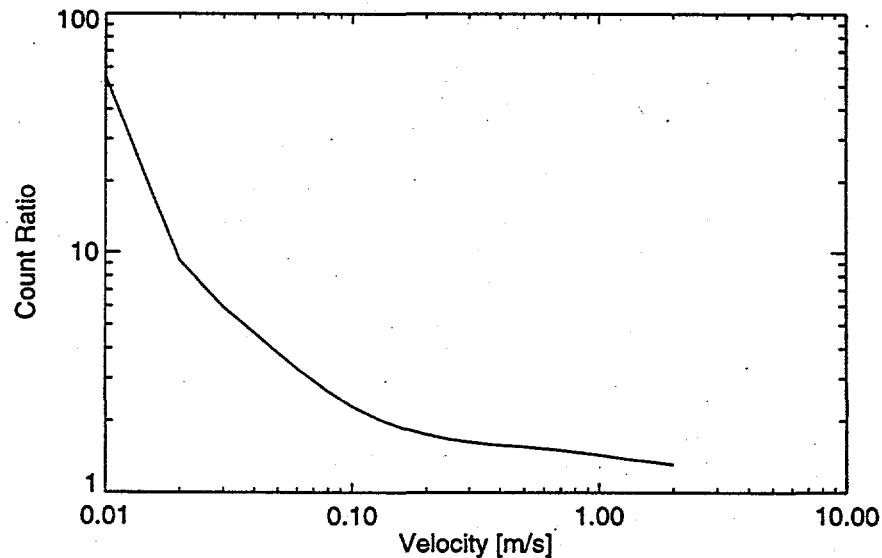


Figure 8. Dependence of Count Ratio on Flow Speed for Detectors Located 3 and 6 m Downstream from Irradiation Source

Conceptual Deployment

The anticipated deployment uses neutron detectors on at least two blending gas streams (enriched input and product output) to detect the delayed neutrons at a distance of 3 m downstream from an irradiation source of ^{252}Cf . Locating additional detectors upstream along the flow lines provides a measure of the background. Measuring and subtracting background is important, particularly because the anticipated signal is significantly smaller than the background.

A single neutron detector downstream from the irradiation source provides a signal that depends on a combination of the ^{235}U content and flow speed. Adding a second detector at a different downstream distance gives additional data for calculating the ^{235}U and the flow speed. Figure 9 shows a blending line with two downstream and two upstream (background) detectors, along with an irradiation source.

The data to be acquired are the neutron counts from each detector on a blending line. No time distribution (Figure 5) is obtained because the irradiation and flow of the gas are continuous, reaching a steady-state value for a given ^{235}U content, gas pressure, and flow speed. Anticipated counting times based on the MCNP simulations are about 10^4 s (about 2.8 hours) to provide a measure of ^{235}U with an uncertainty (1 std deviation) of about 5% for the high-enrichment stream and 43% for the low-enrichment stream.

An alternative method for measuring ^{235}U content and flow is counting the number of gamma rays above a certain energy emitted by the fission products as they move downstream following induced fission. Counting only high-energy gamma rays avoids potential problems from plateout of fission products on pipe walls. The high-energy gamma rays originate from short-lived fission products and provide the measure of the induced fission. The gamma-ray detectors would be arranged similar to the neutron detectors shown in Figure 8.

Finally, if the introduction of an irradiation source, such as ^{252}Cf , is precluded by operational limitations, monitoring of neutrons emitted by (α, n) reactions in the UF_6 is still possible. This method will use a single neutron detector array on each blending line, plus an additional neutron detector to monitor background. The (α, n) method provides no measure of flow speed.

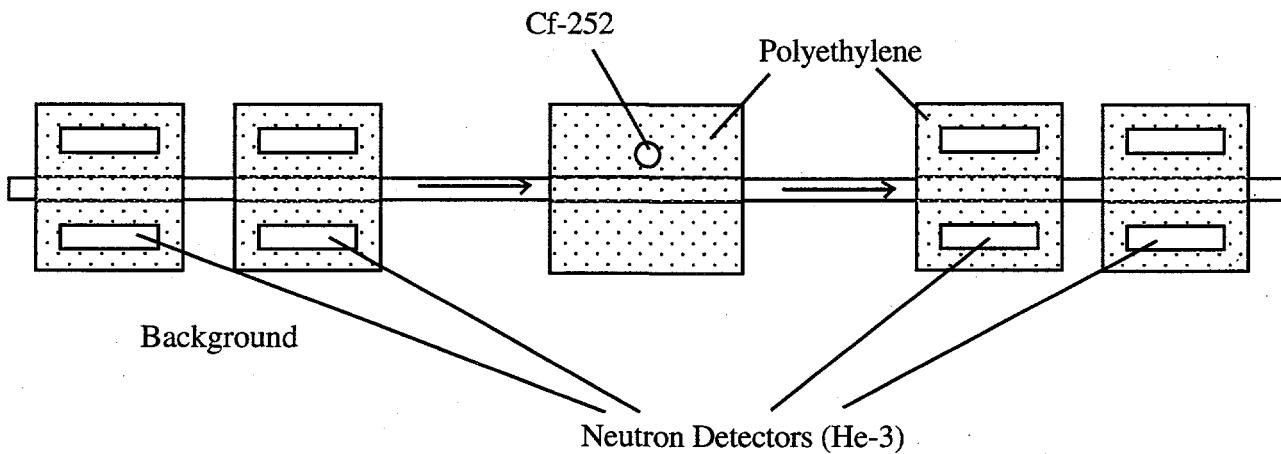
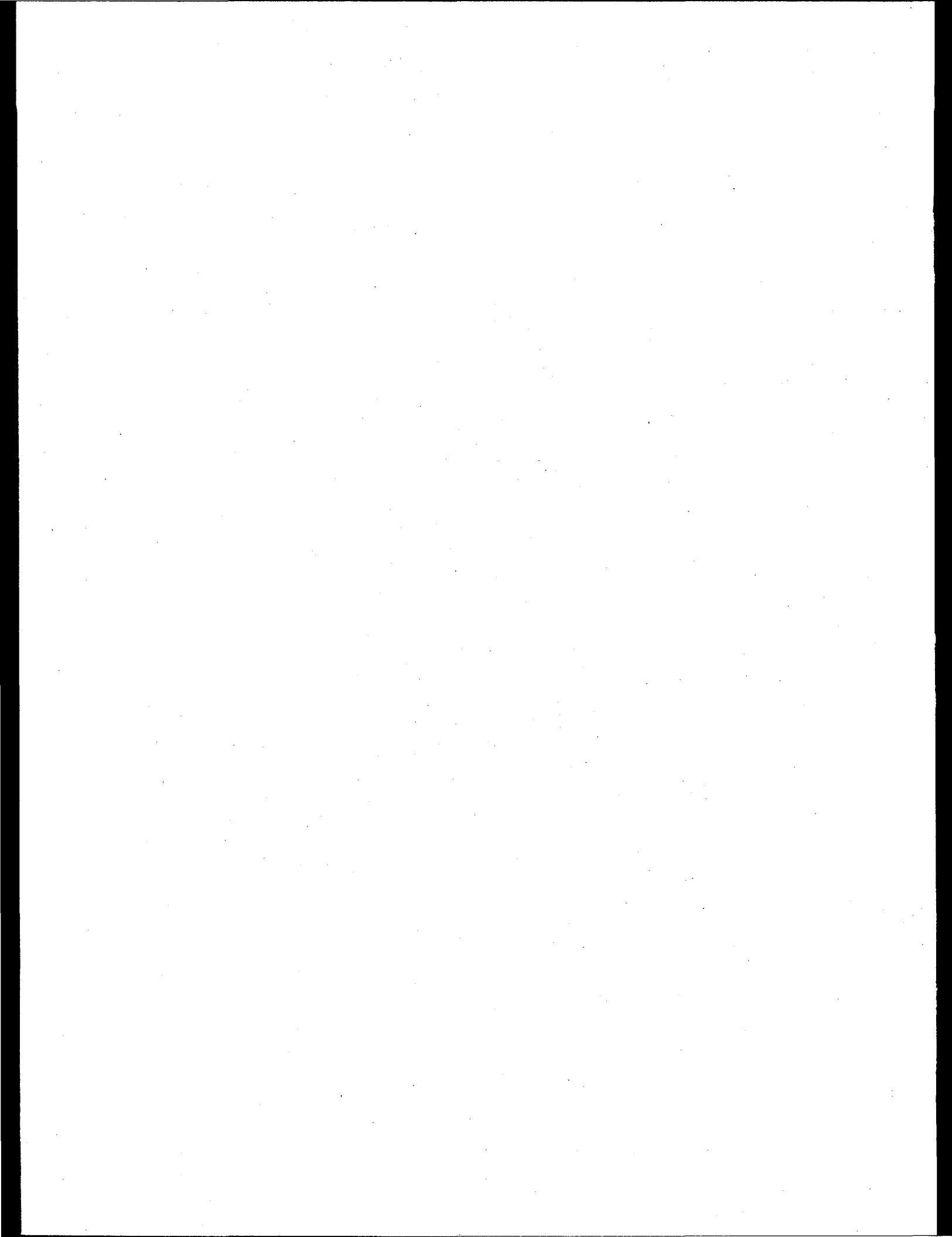


Figure 9. Conceptual Arrangement of Neutron Detectors on Blending Line



Conclusions

Feasibility experiments and computer simulations show that inducing fission in UF_6 gas and measuring the delayed neutrons emitted after the gas and fission products move downstream provides a means for determining ^{235}U content and flow speed. An alternative method measures the high-energy gamma rays emitted from the fission products to obtain the ^{235}U content and flow speed. Monitoring neutrons emitted by (α, n) reactions in the UF_6 gives a measure of ^{235}U without using an irradiation source to induce fission, but no measure of flow speed is obtained.

References

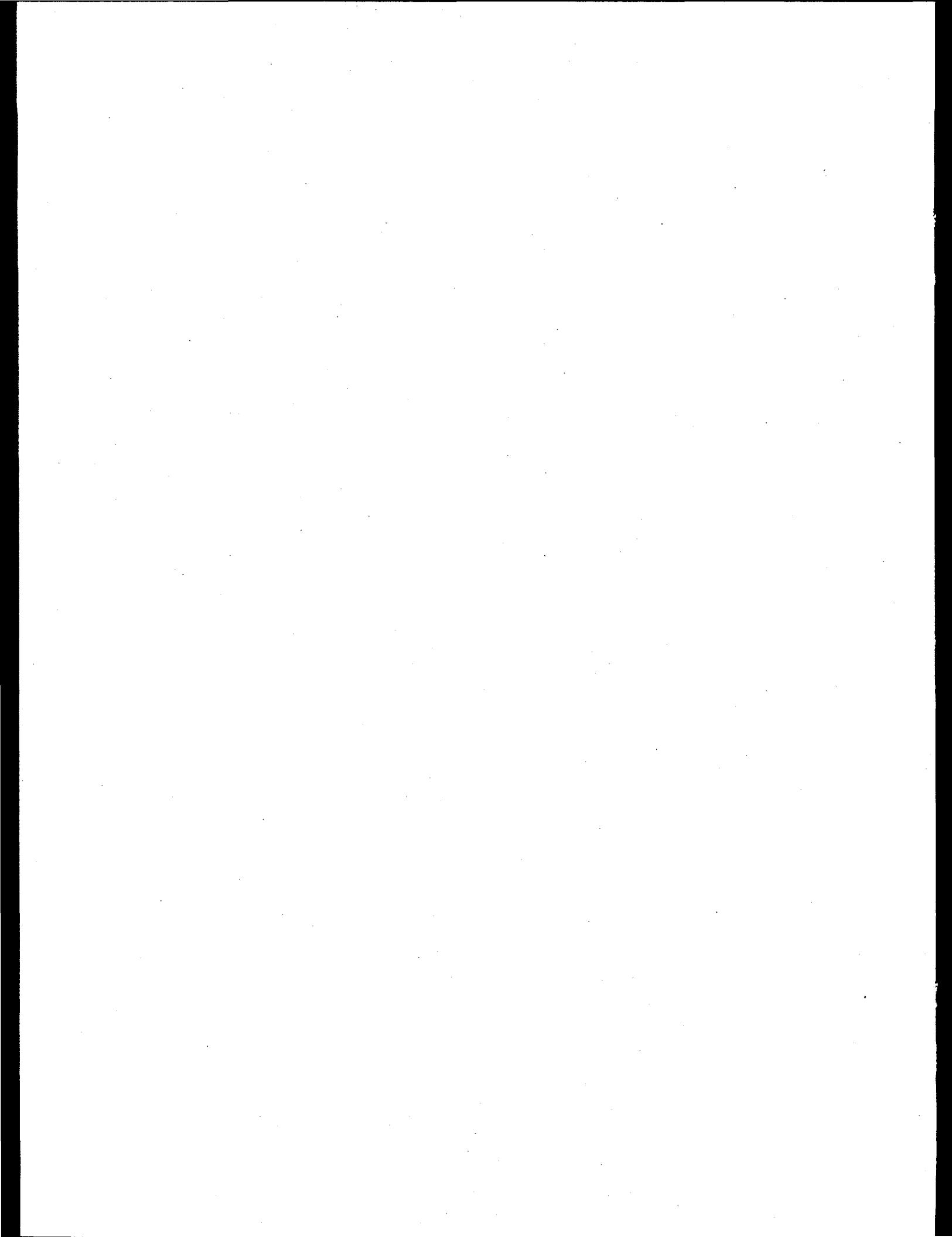
Briesmeister JD (Ed). 1993. *MCNP-- a general Monte Carlo n-particle transport code, version 4A*, LA-12625, Los Alamos National Laboratory, Los Alamos, New Mexico.

Kinsey R (Ed.). 1979. *ENDF/B Summary Documentation*, BNL-NCS-17541 (ENDF-201), 3rd. ed. (ENDF/B-V), National Nuclear Data Center, Brookhaven National Laboratory, Upton, New York.

Northcliffe LC and RF Schilling. 1970. *Nuclear Data Tables* 7A, 233.

Reeder PL and RA Warner. 1984. "Distribution of Delayed Neutron Yields Versus Proton, Neutron, and Mass Numbers: Application to Proton Pairing in Fission Yields," *Nucl. Sci. Eng.* 87, 181.

Reeder PL, JF Wright, and LJ Alquist. 1977. "Delayed-Neutron Emission Probabilities of Separated Isotopes of Br, Rb, I, and Cs," *Phys. Rev. C* 15, 2108.



Distribution

No. of Copies

Offsite

2 DOE Office of Scientific and Technical Information

6 Department of Energy
Attn. M O'Connell, NN-20
Attn. R Malenfont, NN-20
Attn. D Spears, NN-20
Attn. A Bieniawski, NN-42
Attn. R Cherry, NN-44
Attn. D. Dougherty, NN-42
1000 Independence Ave., SW
Washington, DC 20585

1 Oak Ridge National Laboratory
Attn. JT Mihalczo
P.O. Box 2008
Oak Ridge, TN 37831-6004

Onsite

24 *Pacific Northwest National Laboratory*

M Bliss
GB Dudder (5)
HS Miley
RW Perkins
AJ Peurung
PL Reeder
DE Robertson
AJ Schilk
DC Stromswold (5)
RA Warner
NA Wogman

Technical Report Files (5)

