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# Pacific Northwest National Laboratory

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## Neutron Experiments at Portsmouth for Measuring Flow and $^{235}\text{U}$ Content in $\text{UF}_6$ Gas

D.C. Stromswold  
P.L. Reeder  
A.J. Peurrung

April 1997

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

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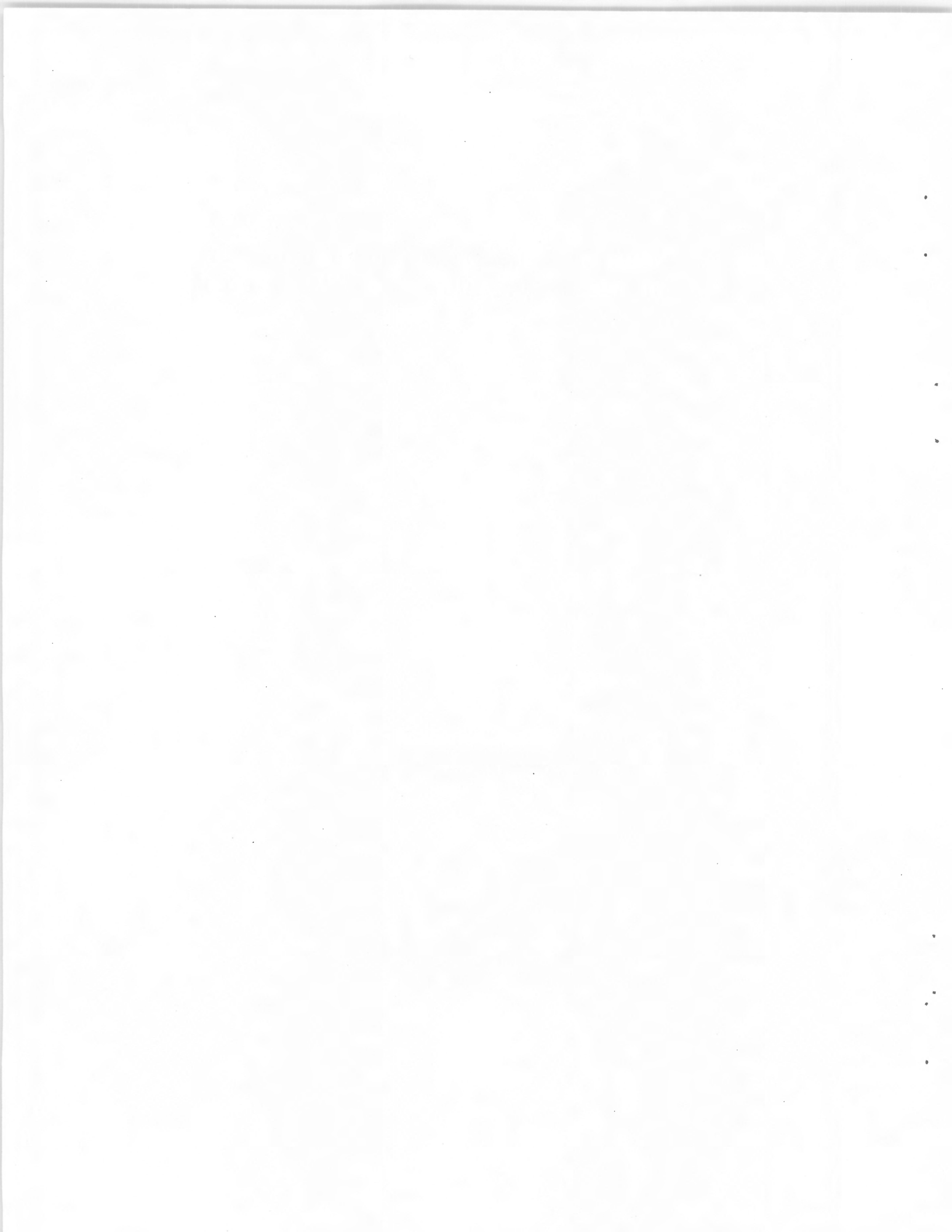
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Richland, Washington 99352



## Summary

The Portsmouth Gaseous Diffusion Plant, located near Portsmouth, Ohio, produces enriched uranium for use in commercial power reactors. The plant also aids disposal of excess high-enrichment uranium (HEU) by blending it with lower-enrichment material. This HEU is fed into the normal product stream as  $UF_6$  gas during this blending process. Methods for non-destructive assay of the material during blending are being investigated in support of possible future International Atomic Energy Agency monitoring of the blending process.

Experiments were conducted in December, 1996, at the Portsmouth Gaseous Diffusion Plant to test two neutron-based methods for monitoring the down-blending of HEU. A goal of the experiments was to conduct tests under realistic plant operating conditions, including temperatures of  $60^\circ$  to  $70^\circ C$  ( $140^\circ$  to  $160^\circ F$ ). The equipment for the experiments was set up on two pipes carrying  $UF_6$  gas being blended as part of the program to reduce the inventory of excess HEU. Results of the initial experiments showed that gas (on-off) could be detected, but that additional tests and data are needed to quantify the flow velocity and  $^{235}U$  content. Future work needs to accommodate the dynamic operating conditions in the plant and reduce the neutron background count rate.

The experiments used a  $^{252}Cf$  neutron source to induce fission in a small fraction of the  $^{235}U$  contained in the  $UF_6$  gas. The first method measured the attenuation of neutrons passing through the low-pressure (5 mm Hg)  $UF_6$  gas in a 7.6-cm (3-inch) diameter pipe containing HEU. The concept was based on the fact that some of the thermal neutrons are absorbed by  $^{235}U$ , thus changing the observed count rate. Experimental results did show changes in the count rate as HEU gas was repeatedly removed from the pipe by "valving off" the supply of HEU being blended. However, the changes observed were larger than expected, indicating that more than just the small amount of HEU gas in the pipe was changing during the experiment. Possibly deposits of  $UF_6$  on the walls of the pipe were changing also. Changes in count rates that may be due to possible electronic instabilities in the data-acquisition equipment were later shown to be small compared to the effects observed at Portsmouth.

The second method, tested on a 20-cm (8-inch) diameter pipe where gas pressure was higher (57 mm Hg), used a modulated neutron flux to induce fission in the  $^{235}U$ . Modulation was achieved by moving a neutron source. Fission products drifted downstream with the gas flow, and some of them emitted delayed neutrons near a detector about 3 m downstream. The amplitude of the signal is proportional to the  $^{235}U$  content of the gas, and the phase shift of the signal, compared to the source modulation, gives the flow velocity. Results of the test showed that enhanced shielding was needed for the neutron source. The background counts in the detectors were too high to allow the expected small signal to be observed.

During both experiments, plant monitoring equipment showed that light gases (freon, oxygen, and nitrogen) were present in widely varying amounts, along with the  $UF_6$  gas. These gases may have affected the experimental results, at least to the extent that they replaced  $UF_6$ .

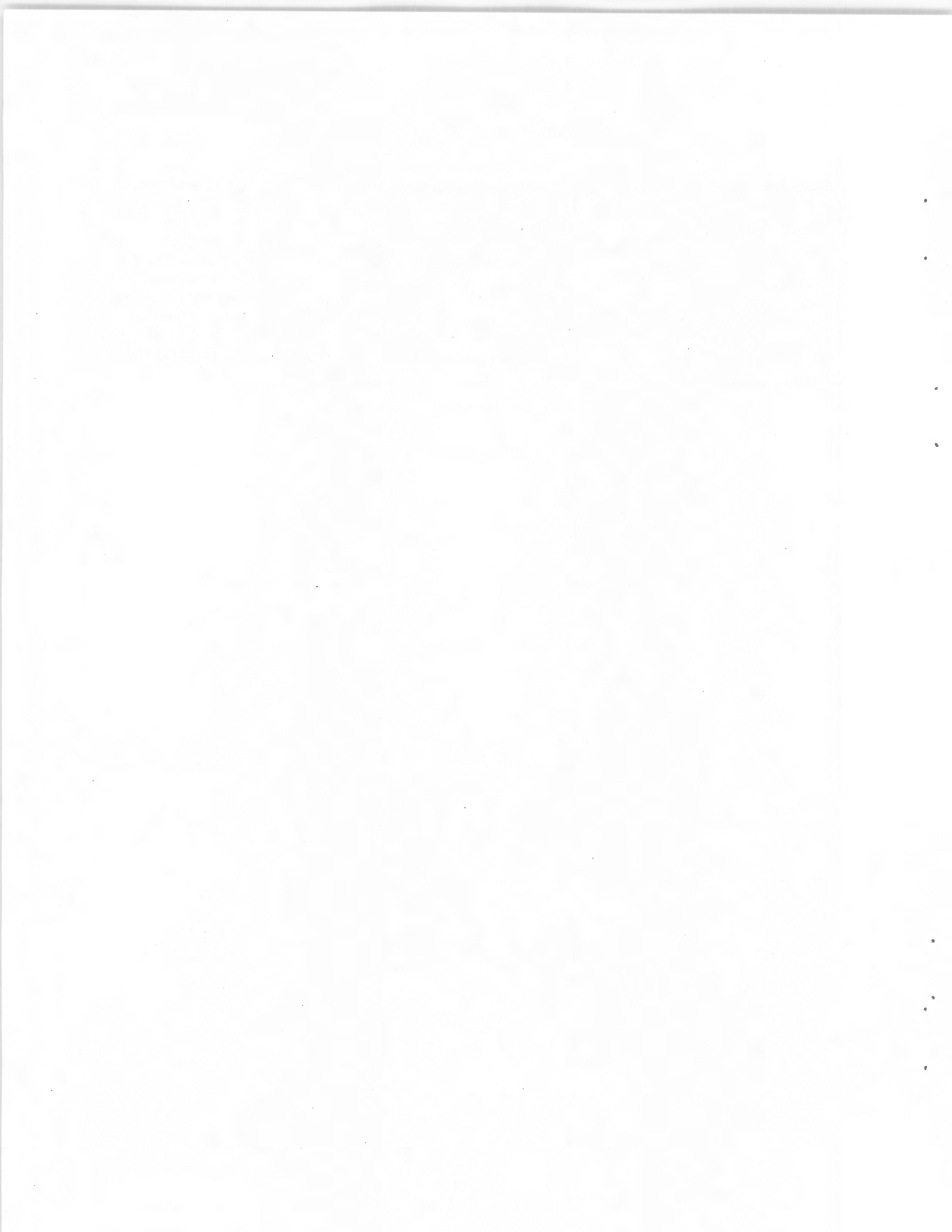
Lessons learned during the initial experiments will be valuable in any follow-on work. These lessons include the following:

- Operating parameters at Portsmouth are dynamic and can introduce significant uncontrolled variables (such as changes in light gases) into the measurements if they are not known.
- Plant operating parameters may vary over short periods (15 minutes) making calibration difficult, and long runs to reduce statistical variations may not be practical.
- Background must be reduced to allow the signal to be observed in the modulated neutron experiment.

This report also contains results of computer simulations and tests performed on the electronics after the experiments were completed at Portsmouth. Recommendations are made for follow-on work to measure the flow velocity and  $^{235}\text{U}$  content.

## **Acknowledgments**

This project was supported by the U.S. Department of Energy, Office of Research and Development, NN-20, under the direction of Michael O'Connell and David Spears and coordinated through the Office of Arms Control and Nonproliferation, International Safeguards Division, NN-440 (Ron Cherry and Amy Whitworth). We would like to sincerely thank Paulette Williams and Eugene Gillispie, DOE/PORTS, for facilitating onsite access and assistance at Portsmouth. David Gordon at Brookhaven National Laboratory (BNL) provided vital assistance in conducting the experiments, as did Ralph Royce of Lockheed Martin Utilities Service (LMUS). The experiment could not have been executed without the cooperation, collaboration, and onsite assistance provided by Lynn Calvert (Lockheed Martin Energy Systems), Bryant Lybrook, Dale Hamilton, Dana Mauk, Richard Mayer, Brent McGinnis, Tom Bonner, Mike Kelley, Buck Sheward, Ken Whittle, David Shisler (LMUS) and Mano Subudhi (BNL).





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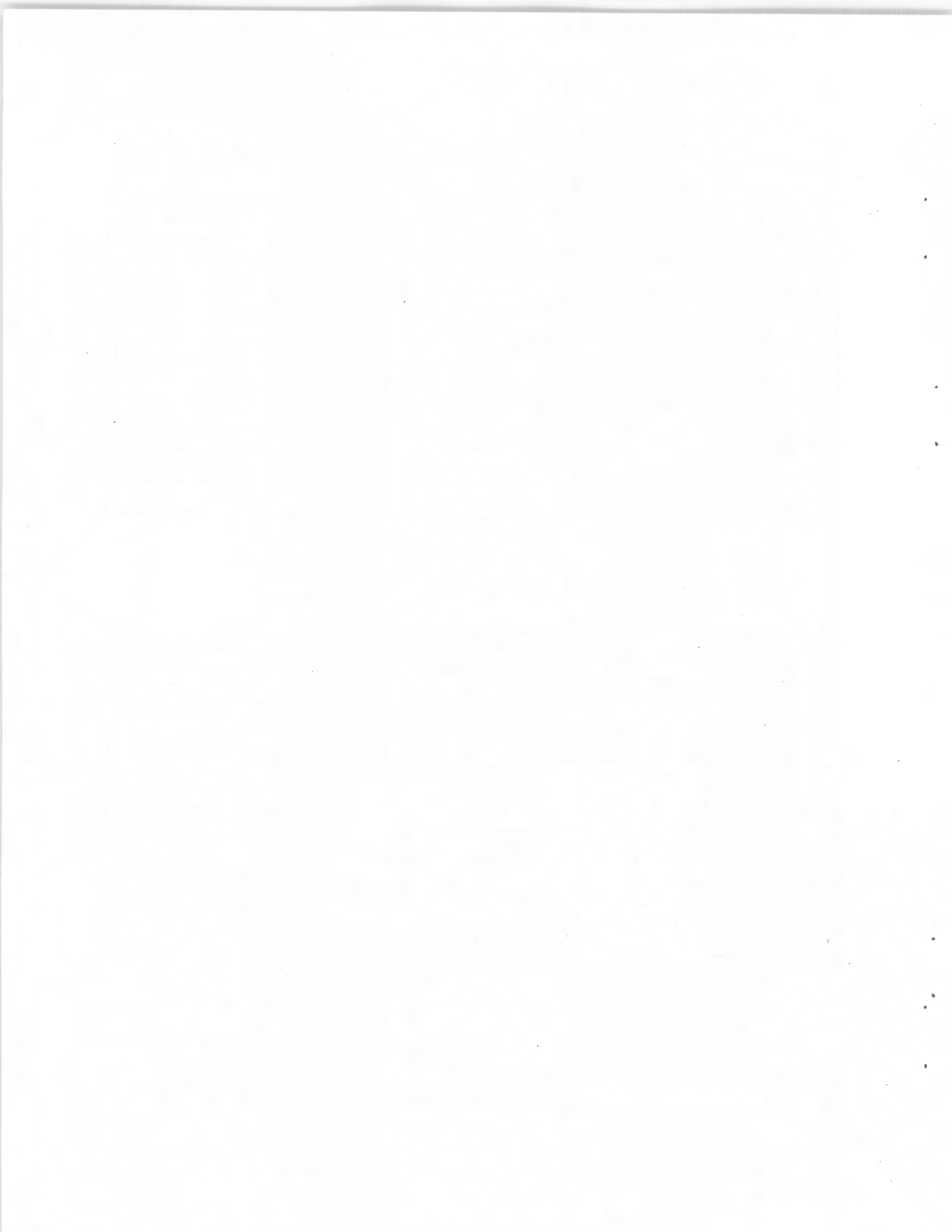
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## 1.0 Introduction

At the Portsmouth Gaseous Diffusion Plant,  $UF_6$  gas streams of highly enriched uranium and lower enrichment uranium are being blended to reduce the stockpile of the highly enriched material. The resultant uranium is no longer useful for weapons, but is suitable as fuel for nuclear reactors. Monitoring the blending at Portsmouth can support International Atomic Energy Agency (IAEA) activities on controlling and reducing enriched uranium stockpiles.

The nuclear properties of the  $UF_6$  gas offer several possible methods for measuring the amount of  $^{235}U$  and its flow velocity. For example, in a technique developed at Los Alamos National Laboratory (LANL), detection of the 186-keV gamma ray emitted by  $^{235}U$  gives a direct measure of that radionuclide. Other LANL techniques include attenuation of gamma rays from a  $^{57}Co$  source as they pass through the  $UF_6$  gas and x-ray fluorescence of uranium. Collimation of the gamma rays reduces the effects from deposits of uranium on the pipe walls. Oak Ridge National Laboratory (Mihalcz 1996) has developed a technique to detect delayed gamma rays emitted following neutron-induced fission of  $^{235}U$ . One of the experiments described in this report is similar to that method, except that delayed neutrons are detected instead of delayed gamma rays.

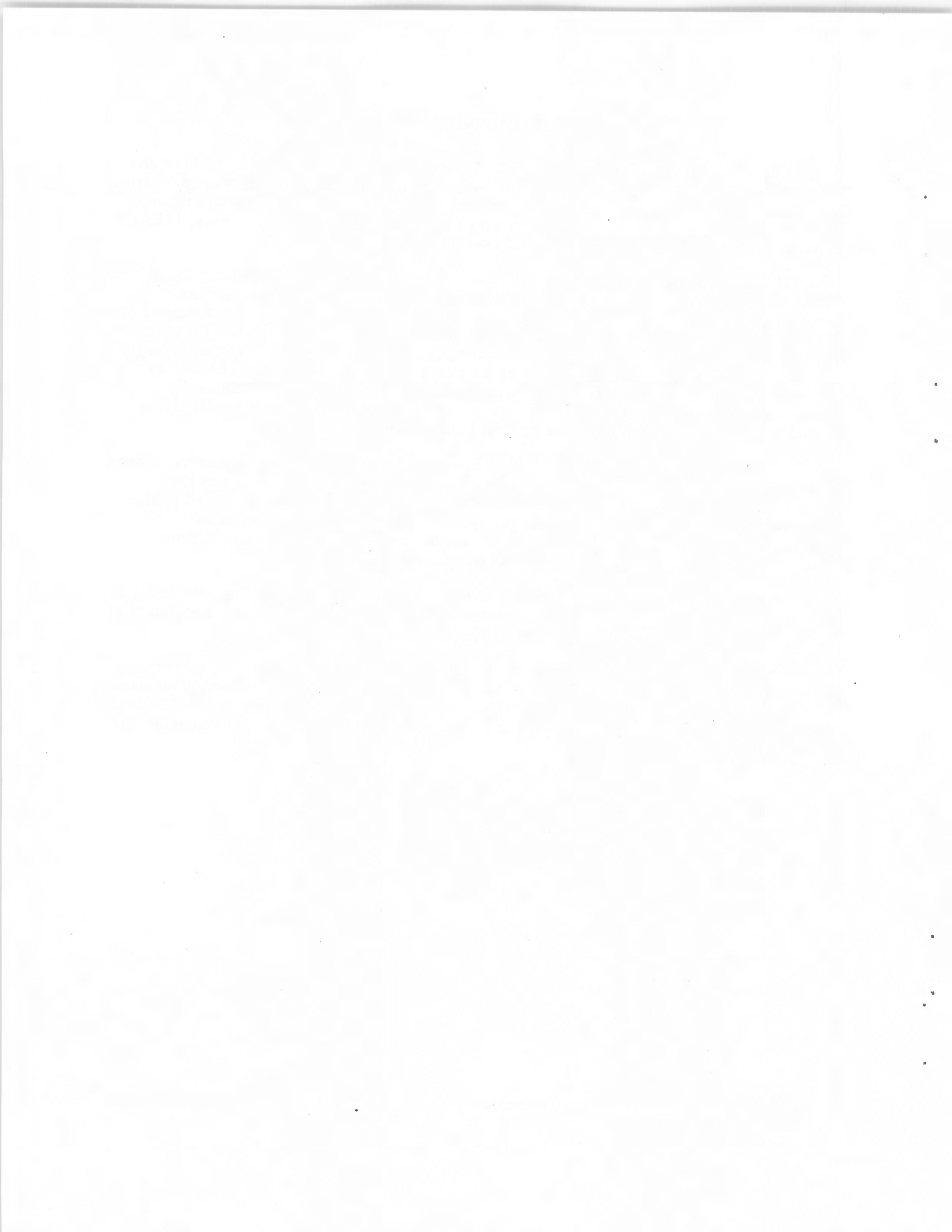
For the Portsmouth application of detecting flowing  $UF_6$  gas, the operating parameters for some of the techniques described above are unfavorable. For example, the gas pressure is very low (5 mm Hg) in the pipe containing high-enrichment uranium. The pipe carrying the mixed product has higher gas pressure (57 mm Hg), but the flow velocity is high (13.5 m/s). The ambient temperature for both pipes is  $> 140^\circ F$ , making the use of some gamma-ray detectors difficult because their light output decreases significantly with increasing temperature.

Two experiments to monitor the blending of high- and low-enrichment uranium were tested at the Portsmouth plant by Pacific Northwest National Laboratory (PNNL)<sup>(a)</sup> for the U.S. Department of Energy, Office of Research and Development (NN-20).

The methods investigated for monitoring the  $^{235}U$  content in the  $UF_6$  gas have the common feature of using a neutron source to induce fission in some of the  $^{235}U$  nuclei. One method measures the attenuation of thermal neutrons crossing a pipe containing  $UF_6$ . A second method monitors the delayed fission neutrons emitted downstream in the flowing gas following fission induced in the  $^{235}U$  by a modulated neutron source.

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



## 2.0 Test Site

The tests took place at the Portsmouth Gaseous Diffusion Plant, which is located near Portsmouth, Ohio. Building X-326 at the Portsmouth plant is the site of high-enrichment uranium (HEU) blending in the United States. Blending was taking place in cell 25-7-7 of the enrichment cascade at the time of the experiments, and that cell was the location for the experiments. If blending had been taking place at cell 25-7-5, its normal location, fewer problems with light gases moving past the monitoring locations would have been encountered.

Figure 2.1 shows schematically the blending operation with the monitoring equipment installed on the HEU feed pipe and the output (blended) pipe. HEU flows from storage cylinders located about 100 m away from the blending location. The pipe through which the HEU flows has a 2.5-cm (1-inch) inside diameter for most of its length, although at some locations the pipe's diameter is 7.6 cm (3 inches). At the location of the attenuation measurement in cell 25-7-7, the diameter of the pipe containing HEU is 7.6 cm. The equipment for measuring the neutron attenuation was located near the door of the cell, approximately 4 m before the blend point.

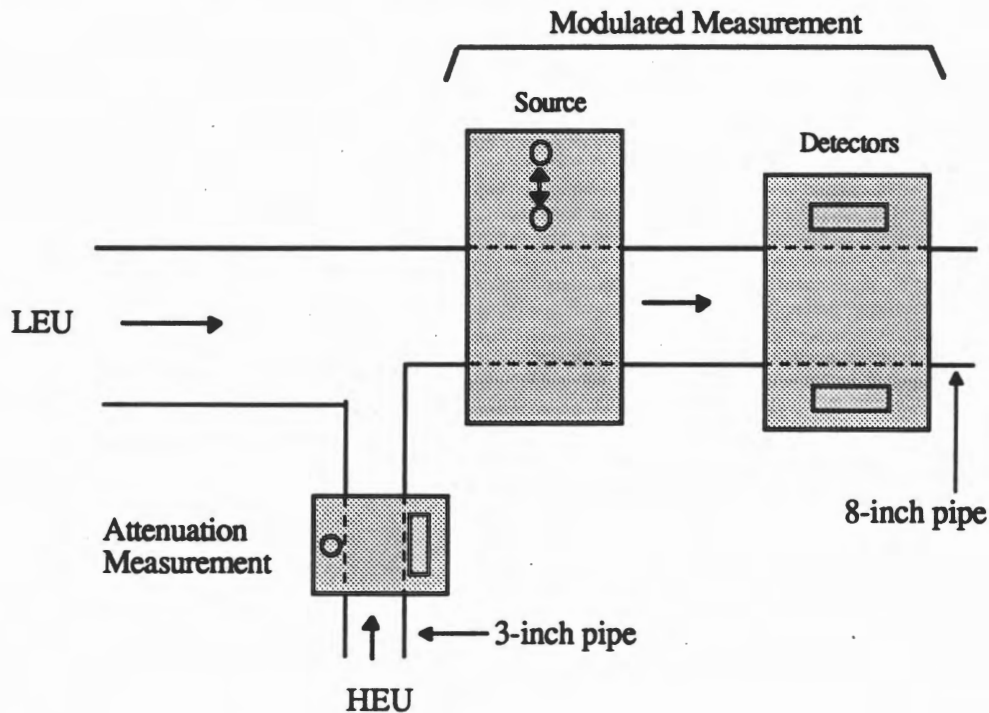


Figure 2.1. Monitoring Equipment on UF<sub>6</sub> Blending Pipes

The equipment for the modulated equipment was installed on the 20-cm (8-inch) diameter pipe that contains the blended UF<sub>6</sub> gas. This pipe is located above the HEU pipe at a distance about 1 m from the floor. The modulated neutron source was located about 3 m downstream from the blend point. The detectors for the delayed neutrons were located an additional 3 m downstream from the modulated source.

Table 2.1 shows the UF<sub>6</sub> gas pressures, flow velocities, and pipe diameters at the monitoring locations. The flow velocities are based on the average consumption of HEU (100 lb/day) during blending and the average production of low-enriched uranium (LEU) (70,000 lb/day) at the plant. The flow velocities in the table are approximately twice the values expected before the experiment, and the revised velocities result from enhanced feed rate of the HEU and increased plant production of LEU.

Table 2.1. UF<sub>6</sub> Gas Parameters at Monitoring Locations

	Enrichment (%)	Pressure (mm Hg)	Pipe ID	Flow (m/s)
HEU	20 to 97	5	7.6 cm (3 in.)	1.4
LEU	4 to 6	57	20 cm (8 in.)	13.5

### 3.0 Thermal Neutron Attenuation Experiment

The experiment to measure the  $^{235}\text{U}$  content of the HEU gas took place on the 7.6-cm (3-inch) pipe. Figure 3.1 shows the experimental configuration with a neutron source located on one side of the pipe and a neutron detector on the other side. Polyethylene around the pipe, source, and detector moderates the high-energy neutrons down to thermal energy. Absorption of some of the thermal neutrons by  $^{235}\text{U}$  causes an attenuation in the number of neutrons reaching the detector. Cadmium shielding around the detector, except in the direction toward the pipe, reduces the background from neutrons not coming from the direction of the HEU in the pipe. Secondary neutrons produced as a result of the fission of  $^{235}\text{U}$  do not completely offset the absorbed neutrons because these neutrons are energetic and mainly leave the vicinity of the detector. The count rate in the detector decreases with increasing  $^{235}\text{U}$  content.

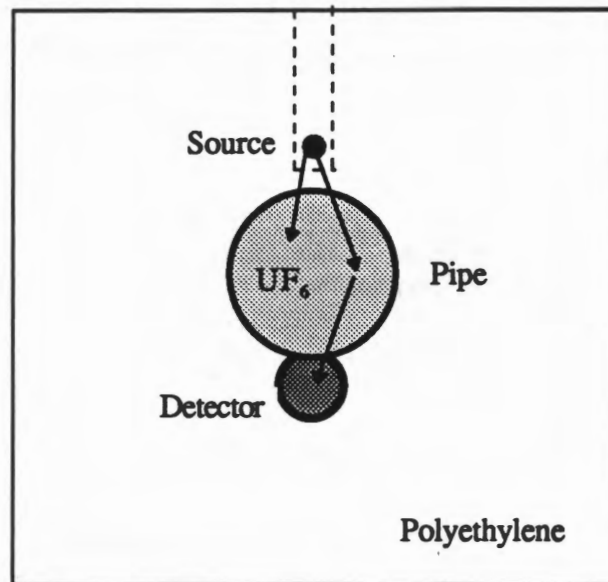


Figure 3.1. Attenuation of Neutrons Passing Through  $\text{UF}_6$  Gas in Pipe

#### 3.1 Equipment

Figure 3.2 is a diagram of the electronic equipment used in the neutron attenuation experiment. As shown in the figure, signals from the detector were amplified, discriminated, and counted in scalars, with output being recorded in a computer. The discriminator removed any contribution to the signal from gamma rays.

The neutron source was located about 10 cm (4 inches) above the pipe in the polyethylene. Tests of other distances for the source from the detector, obtained using polyethylene plugs in the source hole, showed that the 10-cm position gave a sufficiently large count rate (13,500 c/s) to gather data promptly, but not overload the counting system. The neutron detector was located in a slot in the polyethylene, immediately below the pipe, as shown previously in Figure 3.1

The polyethylene moderator was assembled in pieces that fit closely around the pipe. An aluminum platform supported the polyethylene at a distance of about 10 cm (4 inches) above the floor. Details of the neutron source, detector, and polyethylene shielding are as follows:

Neutron Source:  $^{252}\text{Cf}$ , 5.7  $\mu\text{g}$ ,  $1.3 \times 10^7$  n/s  
 Detector:  $^3\text{He}$ , 4 atm., diameter 2.5 cm (1 inch), length 36 cm (14 inches), manufacturer: LND  
 Polyethylene: 46 x 46 x 55 cm (18 x 18 x 21.5 inches)

During the attenuation experiments, the equipment for the modulated experiment (including the polyethylene and motor for moving the source) was not located in the cell with the attenuation equipment.

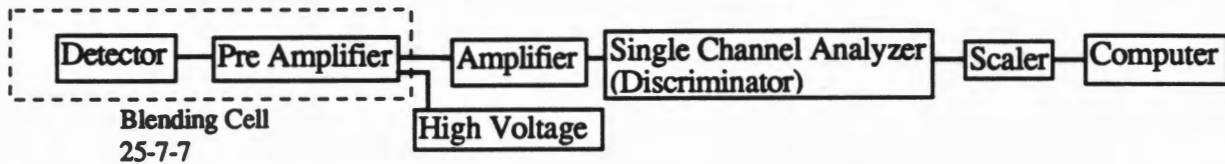


Figure 3.2. Electronic Equipment for Attenuation Experiment

### 3.2 Data Collection

Valving the HEU re-feed cylinders on and off at intervals of about 3 hours allowed data collection when HEU was present and absent in the pipe. The gas was "off" for only two intervals to minimize the time unavailable for HEU blending in the plant. The pressure of the HEU gas was approximately 5 mm Hg when the HEU was present. Several cylinders were being fed simultaneously at the time of the experiment. The exact enrichment of the HEU was unknown, but it was greater than 90 percent.

Most of the data were collected and stored in 500-s time intervals so that short-term changes in individual sets of data could be isolated during analysis in case the dynamic operating conditions in the cascade affected the results.

While the neutron attenuation data were being collected, the output of plant sensors for freon, oxygen, and nitrogen were also recorded at 10-minute intervals. Calibration factors for these sensors are not available, but changes in the magnitudes of the values indicate times when the amounts of these gases were high. During the attenuation experiments, a rupture in equipment containing freon for cooling injected freon into the cascade. This freon "bubble" moved along the cascade and passed through cell 25-7-7, where the neutron equipment was located. When the freon concentration was at its highest, about 90% of the  $\text{UF}_6$  gas was probably displaced by the freon, although the total gas pressure remained approximately constant (private communication from Richard Mayer at Portsmouth). Figure 3.3 shows a plot of the freon during the experiment. Oxygen and nitrogen values also fluctuated during the experiment. These gases can enter the cascade as the result of vacuum leaks or specific operations in the normal functioning of the plant. Figures 3.4 and 3.5 show the measured oxygen and nitrogen values. It is not anticipated that the light gases would enter the measurement region of the HEU feed pipe where the attenuation experiments were conducted, but the data are included for completeness.



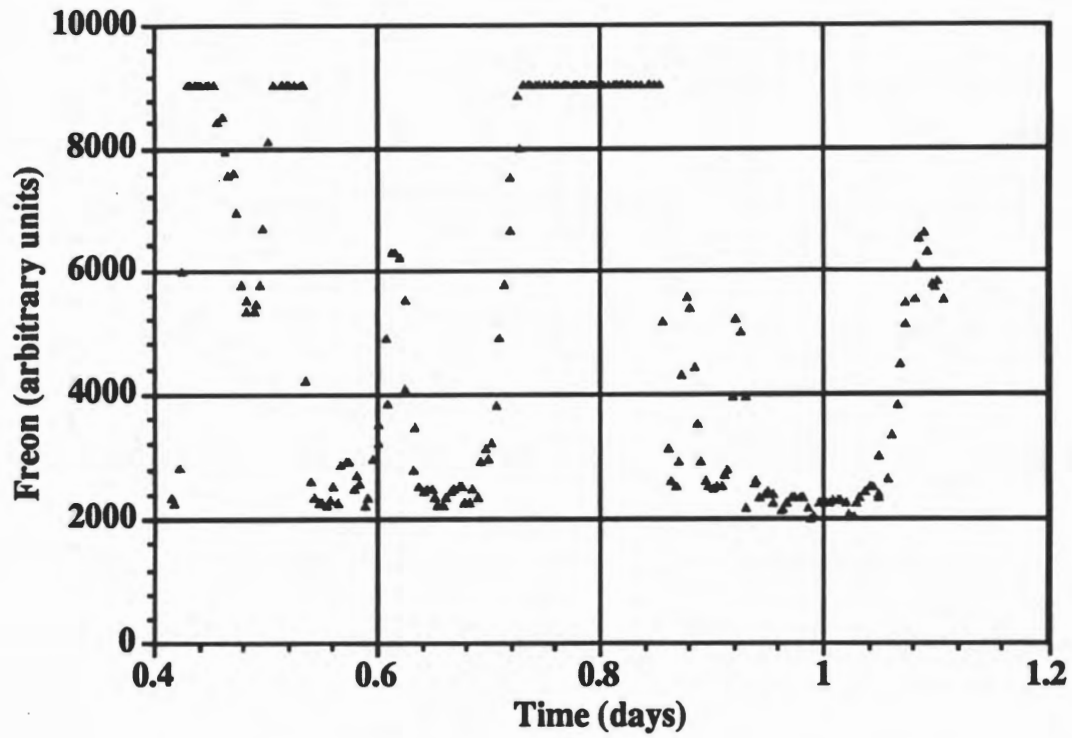


Figure 3.3. Freon Variation During Attenuation Experiment

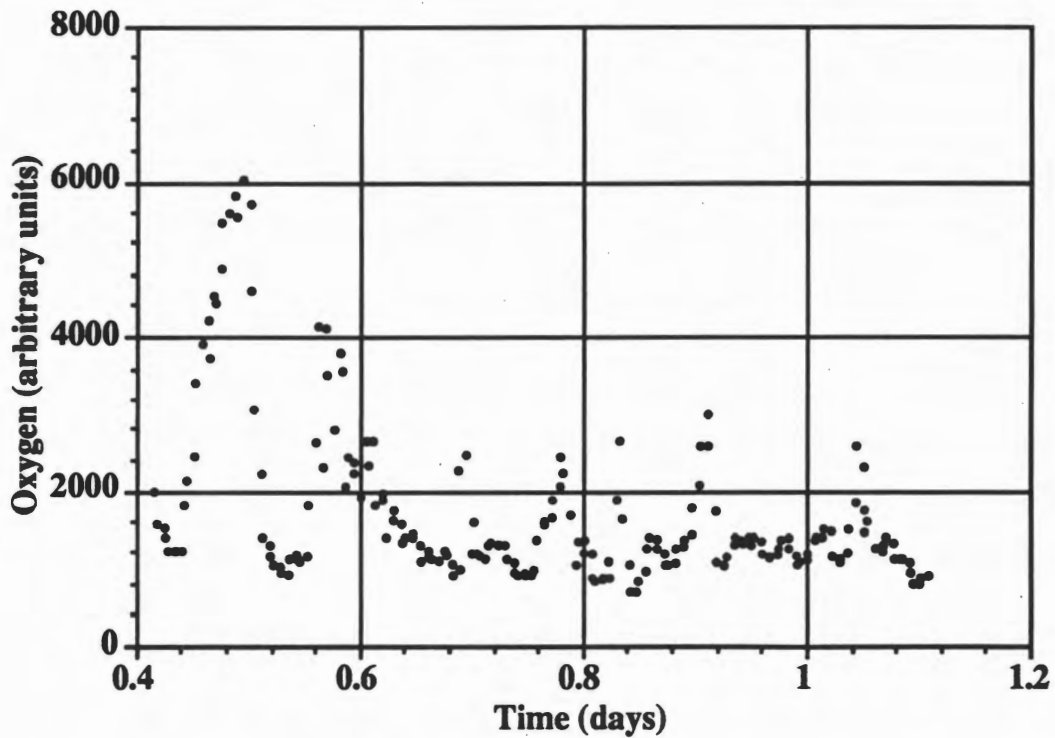


Figure 3.4. Oxygen Variation During Attenuation Experiment

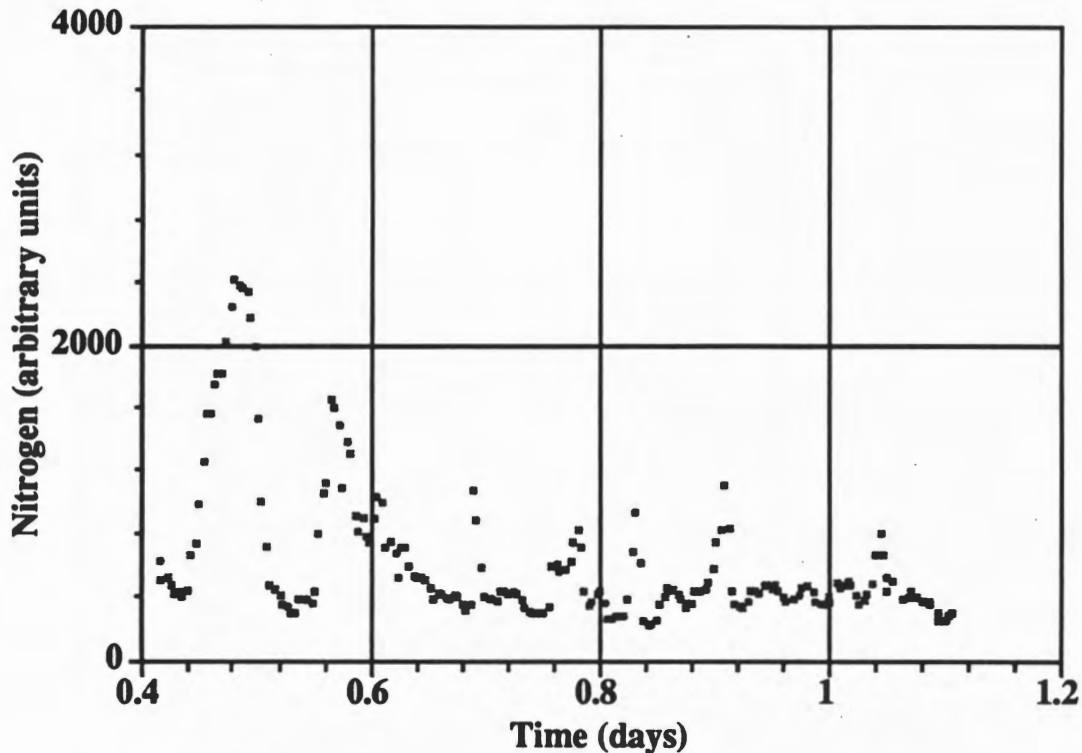


Figure 3.5. Nitrogen Variation During Attenuation Experiment

### 3.3 Results

Figure 3.6 shows the neutron count rates obtained during the attenuation experiment. The figure shows the times that the HEU feed cylinders were valved on and off. Distinct changes in the count rate generally occurred at the times of HEU gas transitions. However, the magnitudes of the changes were not the same each time. For example, a large increase in the count rate occurred at the initial transition to "no gas" in the pipe. When gas was put back in the pipe, the count rate decreased, as expected, but it did not return to the original value. The count rate also changed at subsequent gas transitions, but a trend of generally increasing count rate with time occurred. The large change in count rate at the first removal of HEU gas from the pipe may have originated from the initial disturbance of a long-term operating condition of HEU blending.

Figure 3.6 also shows the expected count rate changes, as determined by computer simulation using the Monte Carlo code MCNP for a transition from HEU at 5 mm Hg pressure to vacuum in the pipe. The expected changes are clearly much smaller than those observed experimentally.

Although the origin of the large observed changes is not known, uncontrolled variables during the experiment may have contributed to the changing count rate. These variables are the uranium coating on the inner wall of the pipe containing the HEU gas, the light gases in the pipe during portions of the experiment, and the temperature of the detector and electronics.

Changes in the amount of uranium on the walls of the pipe could have an effect on the observed neutron count rate because the neutrons can be attenuated both in the HEU gas and in the HEU on the pipe walls. At low gas pressure, such as 5 mm Hg, the amount of HEU gas is small, and comparable amounts of HEU could be attached to the walls. In particular, the amount of  $UF_6$  in the gas would be equivalent to a wall coating thickness of  $4 \times 10^{-5}$  cm inside a 7.6-cm (3-inch) diameter pipe. The likelihood of a change in coating thickness during the experiment is difficult to assess. If

the coating is firmly attached to the walls, such as results from chemical reaction with the pipe surface or moisture-induced chemical reaction, decreases in the thickness seem unlikely.

Physical adsorption of  $UF_6$  molecules to the pipe wall can also occur. Transitions between gas and solid phase occur as indicated in the phase diagram (Figure 3.7) for changes in temperature and pressure. The operating temperature of the cell where the experiment was located is about  $140^\circ F$ , although at the location of the experiment (near the floor and open door), the air temperature could have been as low as  $95^\circ F$ . The temperature measured at a height of 1 m was  $118^\circ F$ . The nominal gas pressure in the pipe is 5 mm Hg (0.1 psi), which is the suction pressure for the nearby pump. The phase diagram clearly shows that the  $UF_6$  will be in the gas phase for this operating temperature and pressure. Changes in  $UF_6$  adsorbed to the walls thus are difficult to explain based on phase change (sublimation or desublimation).

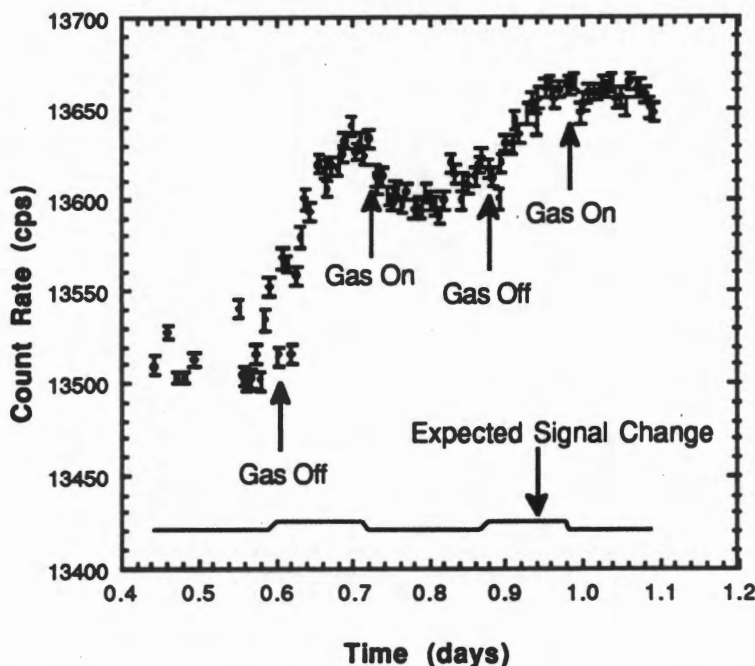


Figure 3.6. Neutron Count Rate in Attenuation Experiment

If light gases (freon, oxygen, and nitrogen) were present in the HEU pipe during portions of the experiment, they would displace the  $UF_6$  gas. Displacement of the  $UF_6$  would cause the absorption of thermal neutrons by the gas in the pipe to decrease because the thermal neutron cross sections for the light gases are significantly less than the thermal neutron cross section of  $^{235}U$  for fission. Comparison of Figures 3.3 through 3.5 for the gases and Figure 3.6 for the neutron count rates shows no obvious correlation between the light gases and the count rates, as expected. For example, the freon abundance changed significantly during the experiment, but the change did not seem to correlate with observed changes in the neutron count rate.

The temperature of the detector and electronics may have changed over the duration of the experiments. Air from a large fan was occasionally directed at the electronics, which were mounted in a rack. The blowing air from the fan may also have entered the cell where the detector was located. Because the fan was not always available, temperature changes may have occurred, but notes

taken during the experiments do not indicate when the fan was present. As described in Section 6.0, experiments conducted after the return of the equipment to PNNL showed count rate drifts attributable to equipment warm-up. Possibly the same temperature sensitivity produced changes at Portsmouth as the temperature changed (either from warm-up or air from the fan).

No mechanism has been positively identified to account for the larger-than-expected changes observed in the neutron count rates during the attenuation experiments. Uranium coating thickness changes on the pipe walls may have occurred, but equilibrium phase transitions between solid and gas should not occur at the temperature and pressure encountered. Although the light gases changed during the experiment, their fluctuations did not correlate with the observed neutron count rates. Equipment malfunction in the neutron detector or electronics is possible, leading to unstable operation. The attenuation equipment located in the cell at the elevated temperature was installed approximately 24 hours before the start of data collection. This time should have allowed the equipment to reach an equilibrium operating temperature, but the temperature of the detector or electronics may have been changed by air from the fan that was used at some times. A longer experiment to monitor changes in count rate would have been desirable, but time was limited. During any future test at Portsmouth, the attenuation equipment should collect data for at least one week to obtain an indication of count-rate changes during normal blending operations.

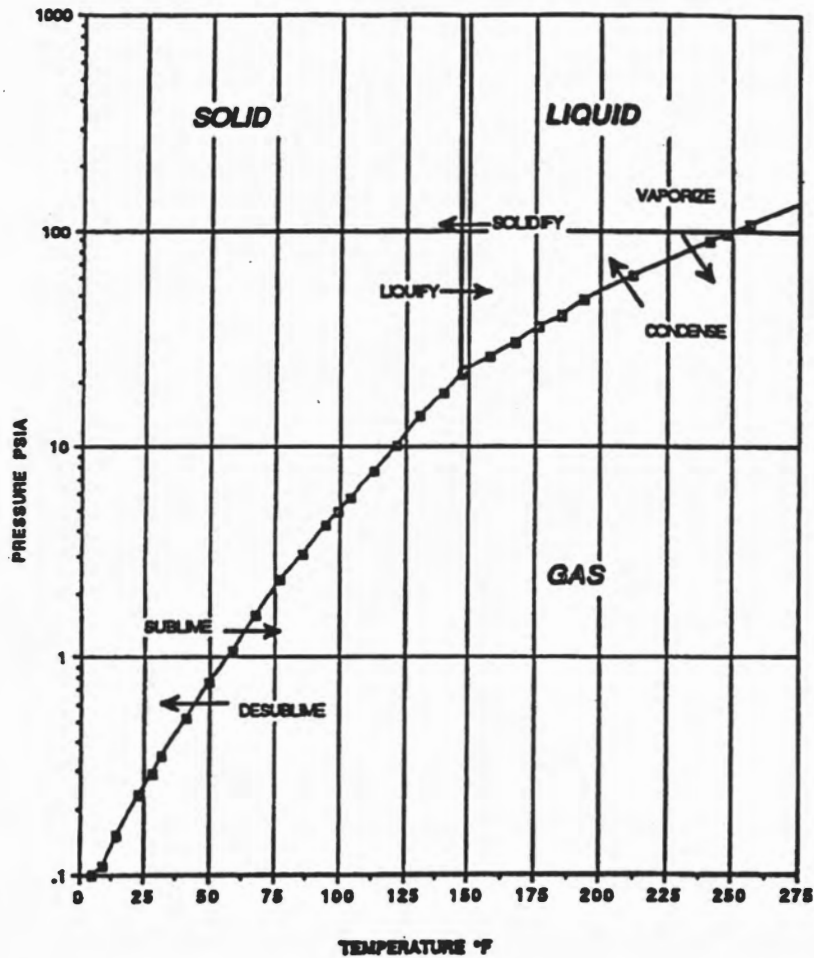


Figure 3.7. Phase Diagram for  $UF_6$

## 4.0 Modulated Neutron Source Experiment

A modulated neutron method was tested on the 20-cm (8-inches) pipe at Portsmouth to determine flow velocity. Moving a neutron source between two positions produces time-varying induced fissions in the  $\text{UF}_6$  gas. The fissions create products that can flow with the gas once the fission products lose their recoil energy, provided they do not hit the walls of the pipe. Some of the fission products that flow with the gas emit delayed neutrons downstream. Detecting these neutrons provides information on the  $^{235}\text{U}$  content of the gas and on the gas-flow velocity. The amplitude of the signal depends on the  $^{235}\text{U}$  content of the gas. The time delay, relative to the source modulation, combined with the travel distance to the source to the detector, gives the flow velocity.

### 4.1 Equipment

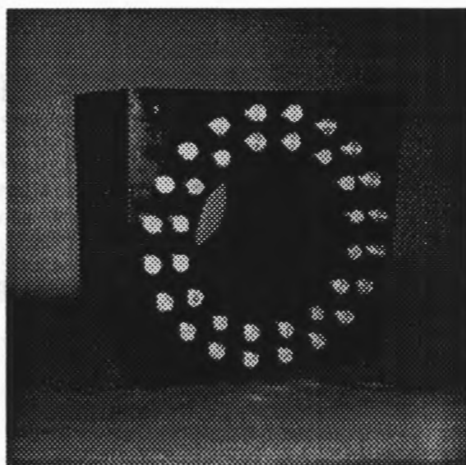
The modulated method requires two main pieces of equipment along the 20-cm (8-inches) pipe through which the blended  $\text{UF}_6$  gas flows: the neutron source, including its motor for movement to produce the modulated signal, and the neutron detectors, which are located downstream at a distance of 3 m. Both the source and the detector are surrounded by polyethylene to reduce the neutron background from the room and from the source. The pipe is about 1 m (39 inches) above and parallel with the floor and has a  $20^\circ$  bend (dogleg) between the source and detector location.

The neutron source (the same one described previously for the attenuation measurement) mounts in a 23-cm (9-inches) arm that is rotated above the pipe by a servo motor. At the "near" position, the source is about 7 cm (2.7 inches) from the pipe. At the "far" position, the source is about 53 cm (20.7 inches) from the pipe. The motor moves the source between the two positions in about 0.04 s under computer-controlled acceleration. Polyethylene shielding around the source was about 46 cm (18 inches) thick, except on the back side (away from the detector) where it was only about 13 cm (5 inches) thick. Cadmium covered a portion of the polyethylene on the side facing the detector. In retrospect, more polyethylene was needed to reduce the background from the source.

An array of 36 neutron detectors embedded in polyethylene and covered on the outside by cadmium was located 3 m downstream along the pipe. Each detector was 2.5 cm (1 inch) in diameter and 36 cm (14 inches) long and filled with 4 atmospheres of  $^3\text{He}$  gas. Figure 4.1 shows the detectors and the inner polyethylene (41 x 41 x 49 cm; 16 x 16 x 19 inches) into which they mounted. The polyethylene was covered by cadmium, and then an additional 5 cm (2 inches) of polyethylene was added around the outside, except on the end facing the neutron source, which had 10 cm (4 inches) of additional polyethylene. The outer polyethylene served as a moderator to slow down energetic neutrons so that the cadmium could absorb them.

The neutron detectors were connected in parallel, with signals going to a single preamplifier, amplifier, and scaler. Because of electrical noise pickup problems, the discriminator for the neutron signal was set high to reject noise. The electrical noise originated from the cable and motor used to move the source. Wrapping the cable and its connectors with copper tape during the experiment reduced the noise to a point where a high discriminator setting could reject it. Reduction of the noise was a major part of the preparation for data collection.

A  $\text{NaI(Tl)}$  gamma ray detector was also used for a small portion of the tests. This detector had a diameter of 12.7 cm (5 inches) and a thickness of 5 cm (2 inches). The detector was located adjacent to the pipe, at the side of the neutron detectors facing the neutron source. No shielding or collimation of the  $\text{NaI}$  detector was used.



**Figure 4.1.** Detectors in Polyethylene for Modulation Experiment

## **4.2 Data Collection**

An Apple Power Book 170 controlled the source movement and the collection of data. Counts from the detectors accumulated in a Camac-system scaler, and they were transferred to the computer upon demand on a periodic time basis to build a time spectrum of counts. Movement of the source from the "far" position to the "near" position started each data cycle, which was repeated many times to accumulate statistically significant data. The data were generally stored in the 100 time bins that comprised a complete cycle of source movement. The individual runs, with their dwell times, number of sweeps (source movement cycles) and other parameters, are listed in Table 4.1. The dwell time per channel was varied from 22.7 ms to 122.7 ms to encompass a range of possible gas flow velocities and corresponding times for the transit of 3 m between the source and the detector. Based on information about the flow obtained before the experiments, the flow velocity was expected to be 6.7 m/s, which gives a transit time of 0.4 s for the gas to move from the source location to the detector. The main data collections (runs 706 to 723 in Table 4.1) were designed to "see" gas moving at this velocity. Other runs listed in the table were performed in case the velocity was slower than expected. Individual runs were kept short (about 10 to 30 minutes) so effects of changes in plant operating conditions (e.g., light gases in the pipe) could be separated later during analysis.

Runs 743 to 750 also collected data from a single NaI detector positioned near the neutron detectors. The discriminator on the output of the detector was set above 2.2 MeV to exclude gamma rays originating from neutron capture by hydrogen. The gamma-ray detector was unshielded, and its brief use did not constitute a valid test of delayed gamma-ray measurements.

The total time of data collection for the modulation experiment was about 16 hours. During this entire time, HEU was flowing for the blending process.

The equipment was set up and tested in a non-operating cell (25-6-7) before conducting the modulation experiment in the blending cell. This allowed the initial installation and testing of the equipment to be made at a lower temperature (about 80° F) and under operating conditions that did not have the special "confined space" designation for personnel entry. Work in that cell provided experience with assembling the equipment in the tight confines of the cell. Experience with data collection tests in this cell also revealed a high neutron background from the <sup>252</sup>Cf source and an electrical noise pickup problem in the neutron counting system due to the motor used to move the neutron source.

Table 4.1. Run Description for Modulated-Source Experiments

Run No.	Bins	Dwell Time (ms)	Irradiate Time (s)	Time span/ sweep (s)	Sweeps
706 to 723	100	22.7	0.27	2.27	3200
724 to 739	100	56.0	0.67	5.6	1575
740 to 742	100	122.7	1.47	12.27	150
743 to 746	50	45.3	0.27	2.26	1300
747 to 750	40	31.7	0.19	1.27	1446

### 4.3 Results

Figures 4.2 through 4.6 show the data collected in the five source-modulation runs listed previously in Table 4.1. In each figure, the higher counts at the start of the time sweep show the time interval during which the neutron source is in the "irradiate" position near the pipe. In this position, the neutrons from the source have higher flux to produce fission of the  $^{235}\text{U}$  in the  $\text{UF}_6$  gas near the source, and the detectors are less well shielded from the source. Many of the neutrons stream down the pipe to reach the detector. The enhanced counts during this time interval are not a problem for data analysis because of the time delay needed for the gas and fission products to drift downstream. Analysis for the delayed neutrons takes place when the source is in the "background" position away from the pipe. The distinct drop in counts when the source moves to the background position is evident in Figures 4.2 through 4.6. An even smaller background count rate is desirable, but it was not achievable with the amount of polyethylene and cadmium available during the experiments.

The expected indication of delayed neutrons is a slightly elevated neutron count ("bump") for a time interval equal to the irradiation pulse width, but delayed by the drift time of the gas (about 0.4 s for the originally expected velocity of 6.7 m/s and 0.2 s for the revised velocity of 13.5 m/s). The solid lines in Figures 4.2 through 4.6 illustrate the magnitude and time dependence of the predicted delayed neutron signal, assuming a magnitude of 0.2 c/s and a drift time of 0.2 s. The "bump" in the solid lines appears at the end of the "irradiate" time and at the beginning of the background time. (The high counts at the end of the irradiation time are artifacts of the data-acquisition system due to movement of the neutron source.) The statistical uncertainties of the experimental data in Figures 4.2 through 4.6 are larger than the expected signal. The background from the neutron source is too large for the expected small increase in counts to appear. A signal of magnitude 0.2 c/s for a flow velocity of 13.5 m/s would be detectable in runs lasting 1 to 3 hours if the background were at the desired rate of 1.5 c/s. However, the actual background is about 21 c/s, which completely obscures any signal from the delayed neutrons. Additional polyethylene and cadmium will reduce the neutron background, and computer simulations (discussed later) give indications of the background reductions to be expected.

Gas velocities greater than about 10 m/s would have been missed in the data collection for the source modulation frequencies used even if the neutron background had not been so high as to obscure the signal. The electronics used to collect the data were not capable of efficient operation for high-speed gas flow. Alternative equipment that is suitable for measuring faster flow velocities without significant data loss is available for future use if the experiment is conducted again.

Gases other than  $\text{UF}_6$  in the pipe during the modulated experiment can also affect the results. Light gases, such as freon, oxygen, and nitrogen, would decrease the expected signal as they displace the  $\text{UF}_6$  because less  $^{235}\text{U}$  would be present to fission and create delayed neutrons. Figure 4.7 shows the variation of freon, oxygen, and nitrogen during the time period of the modulated experiments. The amounts of these gases fluctuated significantly during the first part of runs 706 through 723, but

these gases remained low during the later part of this run sequence. Near the start of runs 724 through 739 and at the end of runs 747 through 750, the freon content was high (at its maximum gauge value of 8990). The short runs for data collection allow those periods with large amounts of light gases to be excluded from the data to see if the signal improves when the light gases are not abundant. Figure 4.8 shows the modulation data for runs 711 through 723, but the signal from the delayed neutrons is still not evident. Similarly, Figure 4.9 shows the data for runs 730, 731, and 736 through 739, during which light gases were not abundant, but again the signal is not evident.

Figure 4.10 shows the multiscaling data obtained from the NaI gamma-ray detector. Because of the lack of shielding on the detector, any possible signal from fission-product gamma rays is obscured by the high background.

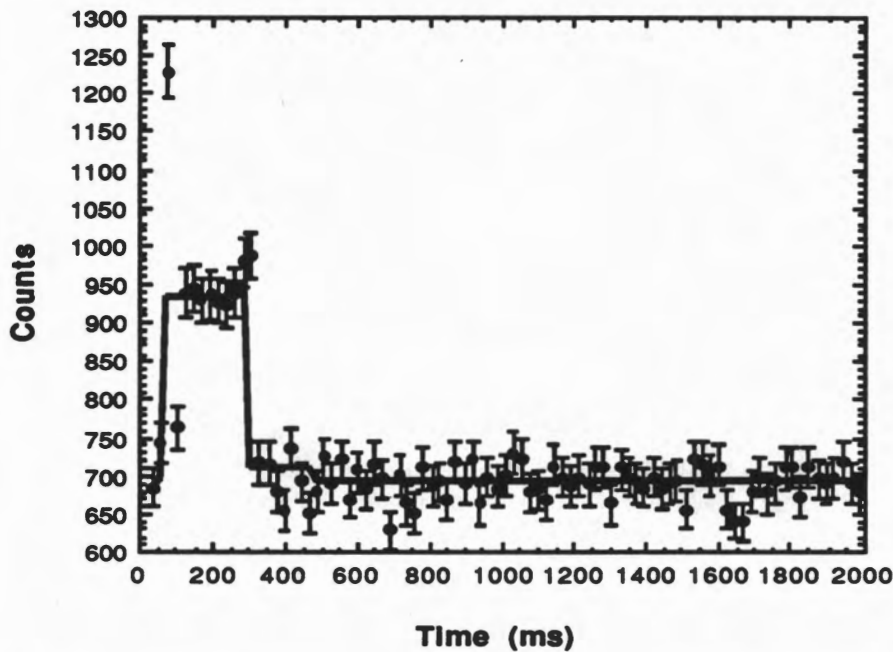


Figure 4.2. Neutron Data from Modulation Experiment (Runs 706-723)



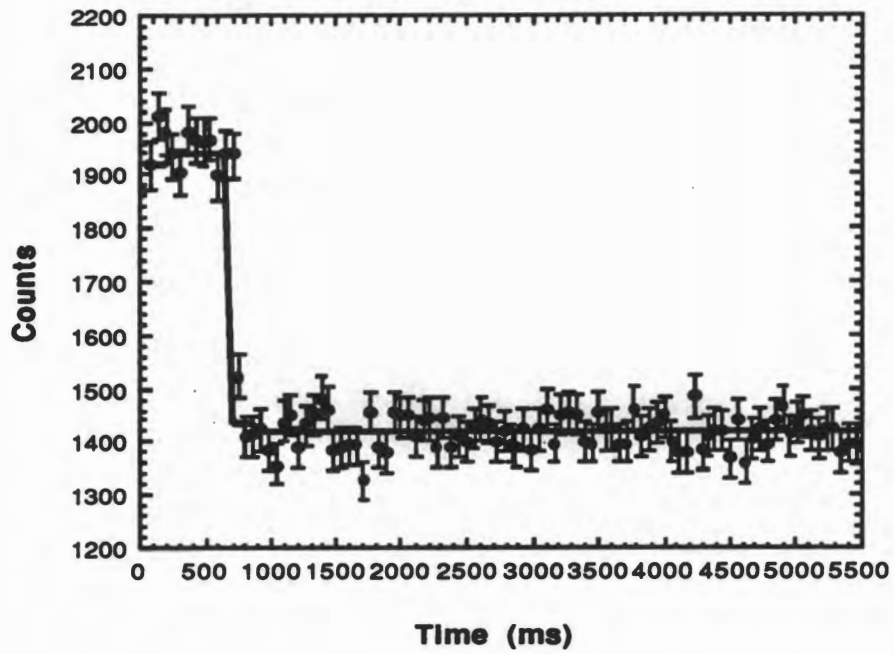


Figure 4.3. Neutron Data from Modulation Experiment (Runs 724-739)

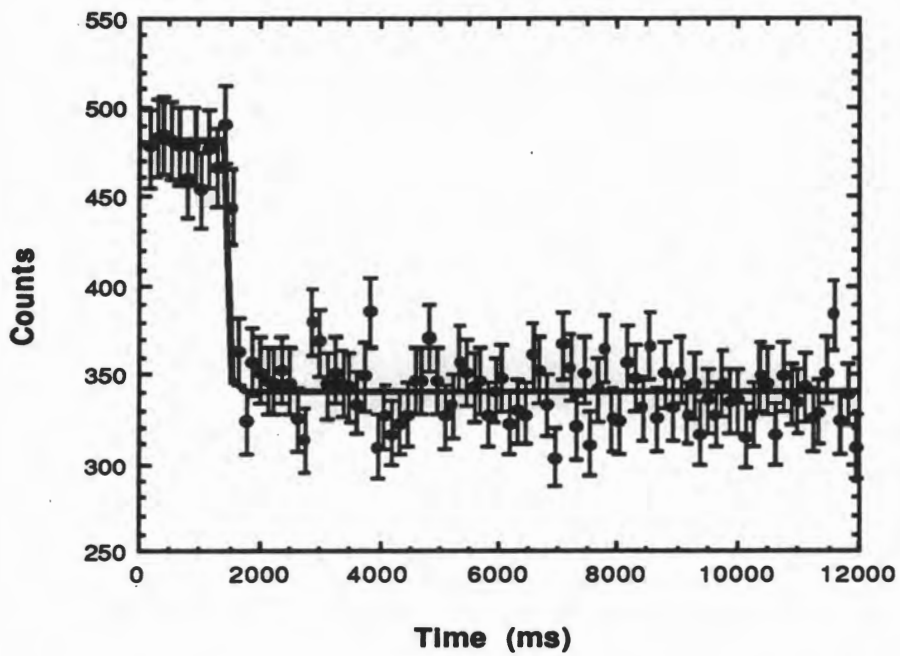


Figure 4.4. Neutron Data from Modulation Experiment (Runs 740-742)

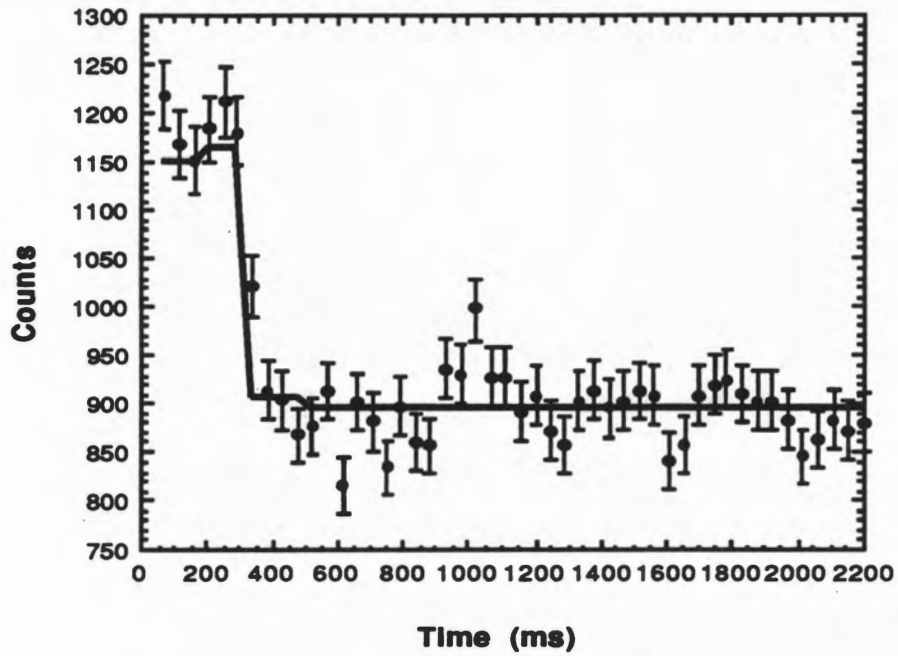


Figure 4.5. Neutron Data from Modulation Experiment (Runs 743-746)

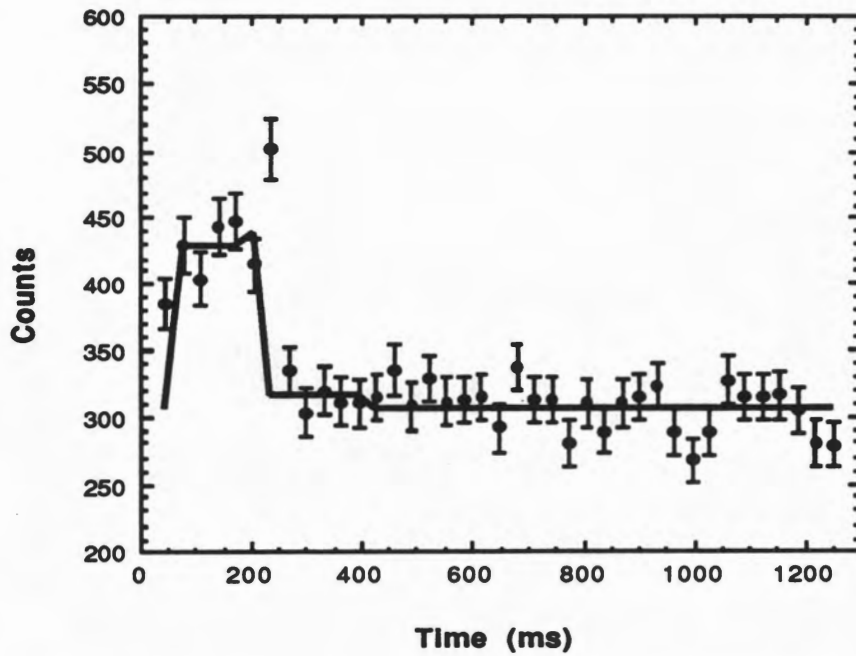


Figure 4.6. Neutron Data from Modulation Experiment (Runs 747-749)

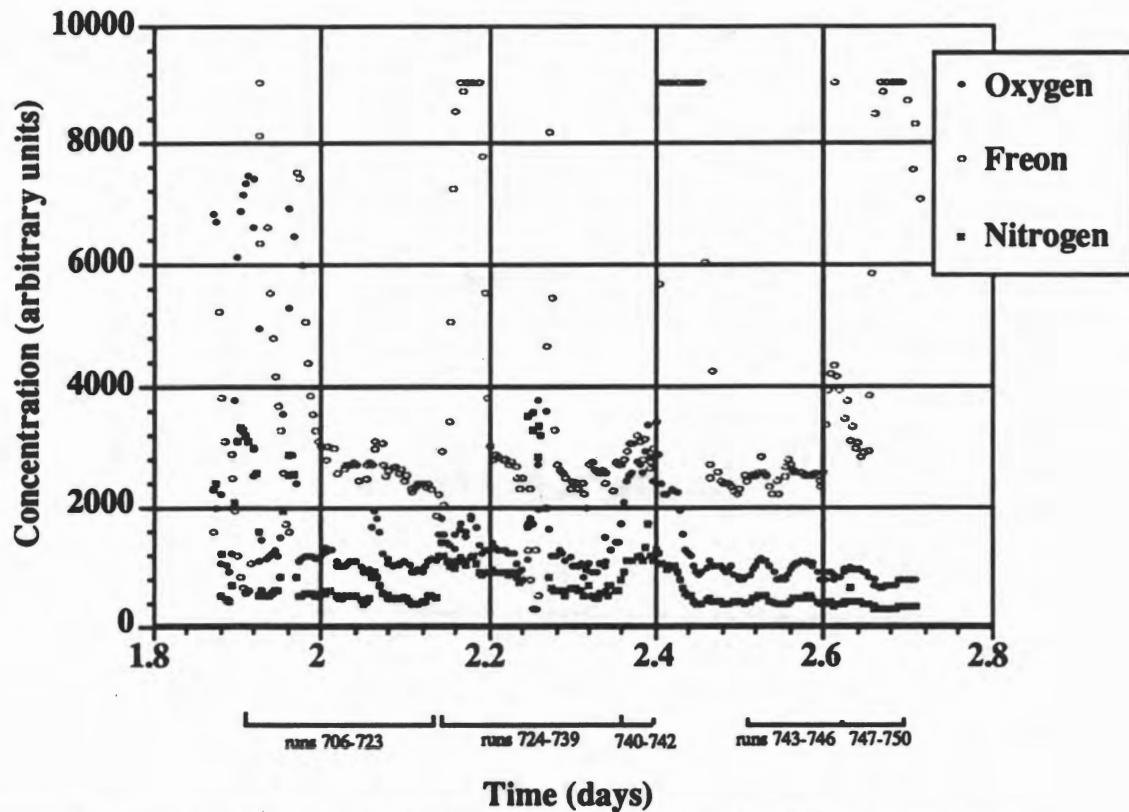


Figure 4.7. Freon, Oxygen, and Nitrogen Variation During Modulation Experiment

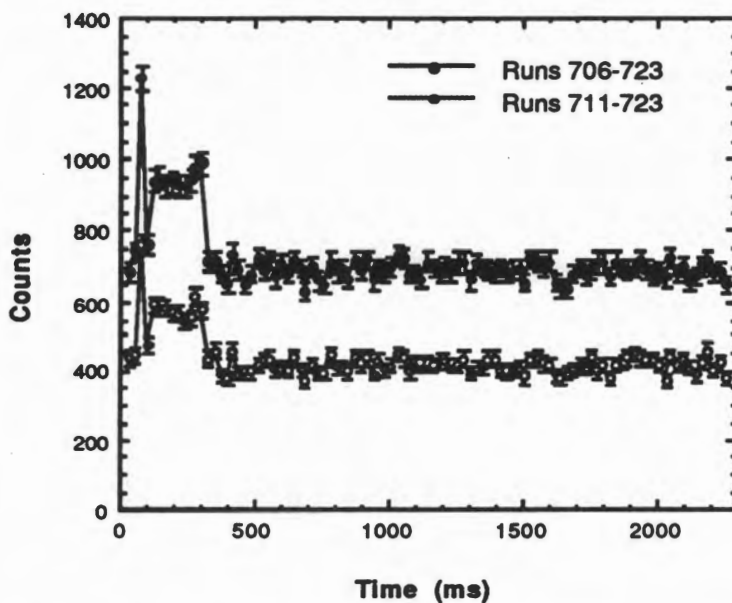


Figure 4.8. Neutron Data from Modulation Experiment (Runs 711-723). Comparison shows data including periods of high light-gas contamination (runs 706-723) and excluding such data (runs 711-723).

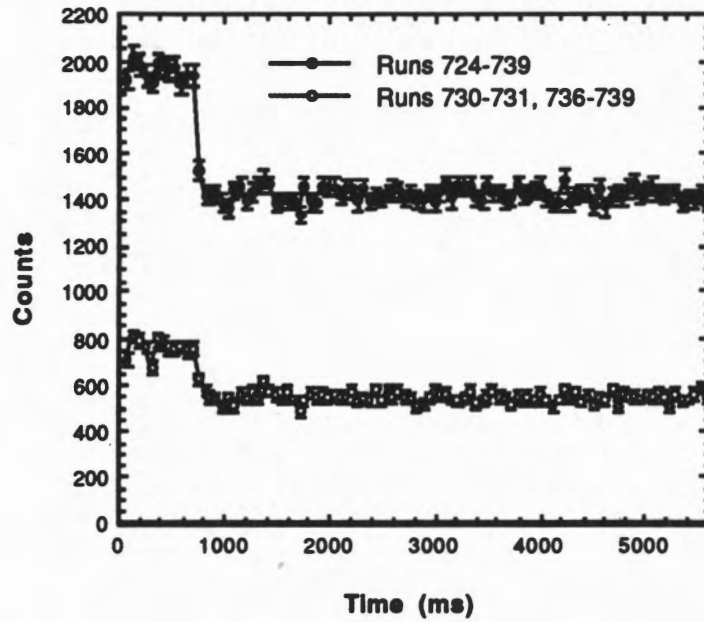


Figure 4.9. Neutron Data from Modulation Experiment (Runs 730, 731, 736-739). Comparison shows data including periods of high light-gas contamination (runs 724-739) and excluding such data (runs 730, 731, 736-739).

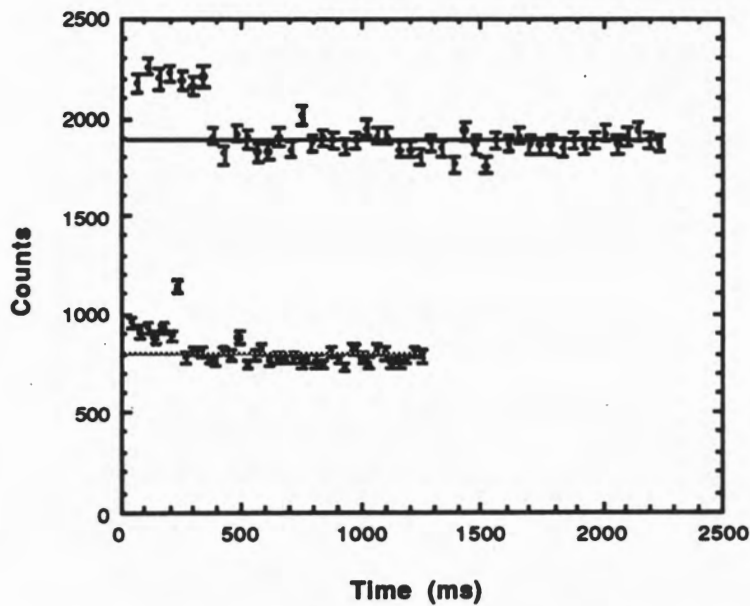


Figure 4.10. Gamma-Ray Data from Modulation Experiment (Runs 743-746 and 747-750)

## 5.0 Computer Simulations

Computer simulations using the code MCNP (Briesmeister 1993) were performed after the experiments at Portsmouth were completed to gain insight on design parameters that could explain observed results or indicate improvements for future experiments. These computer simulations supplement those performed before the experiment and described in previous reports (Stromswold et al. 1996a and 1996b). The simulations did not attempt to reproduce the detailed geometry of the experimental setup. However, the simulations did correctly model the experimental setup with sufficient realism to allow meaningful interpretation of results.

For the modulated method in which the experimental results showed higher than expected neutron background, the computer simulations provided insight on where the main leakage paths were located and how additional shielding could reduce the background. The entire space in some of the simulations was filled with polyethylene, except for the 20-cm (8-inch) pipe. This gave maximum shielding of the detector from the source and showed that essentially no neutrons reached the detector, even through the open pipe. Thus, those neutrons that reach the detector when less polyethylene is present must travel through the air between the source and the detector.

Other simulations used less polyethylene to be more realistic in what can be achieved in practical experiments. Surrounding a bare source with a sphere of polyethylene in the simulations showed the effects of increased source shielding on the neutrons reaching the detector when the source is in the "background" position away from the pipe. For the case of no polyethylene around the source, 0.06% of the neutrons reached the detector region when the source-to-detector spacing, as measured along the pipe, was 2 m. (The 2-m spacing was used rather than 3 m to shorten the computer run time while still providing data on the effect of detector shielding.) The percentage dropped to 0.015% with 10 cm (4 inches) of polyethylene around the source, and to 0.003% with 20 cm (8 inches) of polyethylene.

The 10-cm (4-inch) sphere of polyethylene was used in subsequent calculations to determine neutron scatter from the floor and the benefit of surrounding the pipe with polyethylene. A simulated concrete floor located 1 m below the 20-cm (8-inch) pipe had only minor effect on the number of neutrons reaching the detector. However, surrounding the pipe with 10 cm (4 inches) of polyethylene reduced the number of background neutrons reaching the detector by a factor of 20. Adding cadmium around the polyethylene sphere for the source or around the polyethylene on the pipe had only minor effects on the calculated background neutrons. These results support the conclusion that the background arises from energetic neutrons transported through the air spaces between the source and detector. Polyethylene is more effective in stopping these neutrons than is cadmium. Cadmium absorbs background neutrons that have been thermalized in the polyethylene in the immediate vicinity of the detector.

The simulations also provided the induced fission rate in the  $^{235}\text{UF}_6$  gas when the source was in the "irradiate" position near the 20-cm (8-inch)-diameter pipe. The gas pressure was 100 mm Hg for these calculations, and the  $^{235}\text{U}$  enrichment was 100% in order to expedite the calculations. Scaling the results to the conditions of the 20-cm (8-inch) pipe (gas pressure 57 mm Hg and 5% enrichment) showed that a neutron source emitting  $10^7$  n/s induced fissions at a rate of 2850 fissions/second. The expected signal from the decay of the fission products as they drift downstream can be calculated as follows:

$$Rate = Rate_{IF} \times DN \times DN_d \times Eff \quad (5.1)$$

where

$Rate_{IF}$  = induced fission rate in  $^{235}\text{UF}_6$  gas (fissions/second)

$DN$  = 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

Evaluating the individual terms gives

$$\begin{aligned} Rate &= 2850 \times 0.0167 \times 0.01488 \times 0.30 \\ &= 0.21 \text{ c/s} \end{aligned}$$

Thus the expected signal from the delayed neutrons is 0.21 c/s, based on MCNP computer calculation of the induced fission rate and analytical calculation of the subsequent neutron emission, assuming a source-to-detector separation of 3 m and a flow velocity of 13.5 m/s.

Based on extrapolations of MCNP calculations, the neutron background should be about 20 c/s when the source is in the "background" position (away from the pipe), and about 300 c/s when the source is in the "irradiate" position (near the pipe.) The calculated count rate is in good agreement with the experimentally measured rate (21 c/s) for the background position, but the calculated rate is high compared to the measured rate of 30 c/s for the irradiate position. The MCNP model did not include the 20° bend in the pipe. This should have little effect on the calculated rate with the source in the background position because the fast neutrons from the source do not have a "straight shot" at entering the detector. However, with the source in the irradiate position, the lack of the 20° bend overestimates the calculated rate because it permits "straight shot" access to the detector. The good agreement of the experimental and calculated count rates with the source in the background position gives confidence that the predicted factor of 20 reduction in the count rate by use of additional shielding will in fact be achievable.

Computer simulations were also performed for an alternative geometry of the attenuation method. In this geometry, multiple neutron detectors were located around the 7.6-cm (3-inch) pipe at a distance of 40 cm from the source. This configuration increased the path length of those neutrons traveling from the source to the detector through the gas in the pipe. However, MCNP results showed essentially no contrast in count rate when the pressure of the  $^{235}\text{UF}_6$  gas was changed from 1000 mm Hg to 1 mm Hg. Apparently, the path of the thermal neutrons through the gas does not control the observed count rate. Thus, the original geometry of the attenuation measurement, in which the detector is located directly across the pipe from the source, provides a superior measure of the  $^{235}\text{U}$  content in the pipe.

## 6.0 Experiments at PNNL on Electronic Stability

The equipment for the attenuation was reassembled for stability tests of the electronics and detector after the equipment was returned from Portsmouth to PNNL. A different neutron source was used in these tests because the original source was still at Portsmouth. The PNNL tests showed that a warm-up period of several hours is needed to establish stability for the detection system. The count rates increased over this time period after high voltage was applied to the detector. Figure 6.1 shows an example of the changing counts after application of high voltage. The data are plotted on the same time scale with the Portsmouth attenuation data shown previously in Figure 3.6. Although counts from the stability test increase with time, they do not increase as rapidly or as much as the counts obtained at Portsmouth. Also, the Portsmouth counts did not increase immediately at the start of the experiment, as would be expected from warm-up problems. The main increase at Portsmouth occurred after the first "gas off" transition. An increase in counts at this time, rather than earlier, seems to be associated with gas change rather than electronic drift alone.

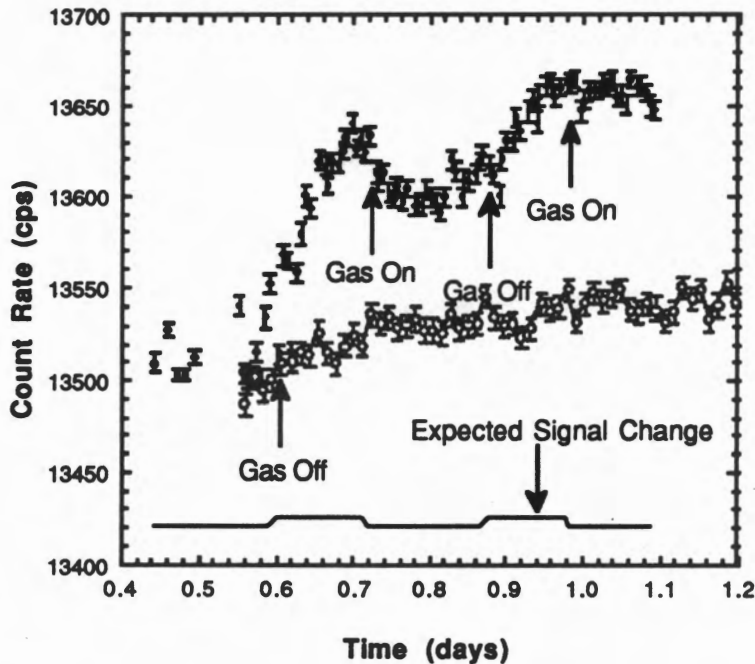


Figure 6.1. Neutron Counter Stability Tests (open circles show stability data at PNNL)

Although the equipment for the modulation experiment was installed in the blending cell on the day preceding the start of collection, the electronics were powered down overnight when no one was present with the equipment. An equipment warm-up period of about 1 hour probably occurred before the start of data collection, based on notes made during the experiment. Equipment warm-up might have contributed to initial instabilities during the attenuation experiment. However, the early data collection periods of the attenuation experiment at Portsmouth (up to the first "gas off" time) showed enhanced scatter, rather than a systematic increase.

For any follow-on experiments at Portsmouth, long equipment warm-up times would need to be allowed before commencing data collection. In addition, collection of data over a longer period than the 1 day used during the initial experiments would provide information on the long-term stability of the measurement. Perhaps the experiment could operate for an extended period in which no HEU is

in the pipe if the blending operation is down for maintenance. In addition, improvements in the electronics would allow operation at higher neutron count rates, which would allow better statistical accuracy in shorter counting times.



## 7.0 Alternative Methods and Recommendations

The desire to provide an acceptable method for the IAEA to use if it chooses to monitor the blending of HEU at Portsmouth is the main motivation for continued development of monitoring methods. The neutron attenuation experiments results indicate that this method should be viable when a more stable data-collection system is implemented. Stability is essential because the expected signal is small, and drifts in the data collection can obscure real changes in  $^{235}\text{U}$  content in the gas.

Gamma-ray techniques, such as those mentioned in the Introduction, could also provide acceptable measurement of the  $^{235}\text{U}$  content. Both Los Alamos National Laboratory (LANL) and the Applied Nuclear Technology Section at Portsmouth are experienced in gamma-ray measurements on  $\text{UF}_6$  to determine  $^{235}\text{U}$ . The low gas pressure in the 7.6-cm (3-inch) pipe may present difficulties in obtaining statistically significant data, as they do with neutron methods. Measurements that require long data-accumulation periods may be precluded at Portsmouth because the gases in the cascade can change over short time intervals, as found during the neutron experiments.

The short-term fluctuations at the plant also present complications to measurements of gas-flow velocity based on neutron or gamma-ray measurements, such as those from modulating a  $^{252}\text{Cf}$  neutron source. Counting delayed neutrons (or gamma rays) requires a low background because the signal measured downstream is weak. Increasing the source strength to boost the signal necessarily results in increased background or the need for additional shielding.

A neutron generator would greatly help in the delayed radiation measurements. The generator would produce an intense pulse of neutrons to increase the signal and then completely shut off to remove background during the subsequent measurement period after the fission products have drifted downstream with the gas. The main disadvantages of a neutron generator are radiation safety, cost, and short operating lifetime (several hundred hours for present generators). The enhanced signal and reduced background will allow measurement of flow and  $^{235}\text{U}$  content in short runs on an intermittent time scale, which greatly extends the operating life of the neutron generator.

A non-nuclear method may provide a quicker and simpler measure of gas flow. However, the inability to insert a probe into the gas stream to measure flow excludes most routine types of measurements. A hot-wire anemometer inserted in the gas stream could measure flow by the dissipation of heat from the probe by the moving gas. An alternative measurement, which requires no probe insertion, is the heating of a small section of the pipe carrying gas. When the gas is not flowing, the temperatures measured at upstream and downstream locations from this heated point should be symmetric. If gas is flowing, some of the heat will be transferred to the gas, resulting in increasing temperature of the pipe in the downstream direction. Figure 7.1 illustrates the pipe heating and temperature profiles. Preliminary calculations indicate that heat transfer to the gas should produce readily measurable temperature changes with gas flow. PNNL has a closed-loop gas flow system that is suitable for testing this method using air flow at various pressures. If the method is successful, then it can be tested at Portsmouth on the 7.6-cm-diameter HEU pipe, where flow can be turned on and off.

### 7.1 Specific Recommendations

The following actions are proposed to provide technology for monitoring HEU blending operations at Portsmouth. The actions are presented in time sequence based on what technologies can be implemented quickly, at least cost, and with maximum likelihood of success.

1. Test the temperature-measurement method for gas flow using an existing closed-loop pipe system at PNNL. The temperature profiles before and after a heated section of pipe can be studied in the lab with and without flowing gas at various pressures. These preliminary tests can be accomplished with minimal cost and elapsed time. If successful in the lab, the technique should

be tested at Portsmouth on the 7.6-cm (3-inch)-diameter HEU pipe as soon as possible. (Note that flow measurement was not attempted previously on this pipe using the modulated neutron technique because the neutron signal was expected to be too small from the low gas pressure.) The temperature-measurement method, in principle, can measure gas flow in a variety of pipe sizes and gas pressures quickly and accurately. The technique does not provide an indication of  $^{235}\text{U}$  content.

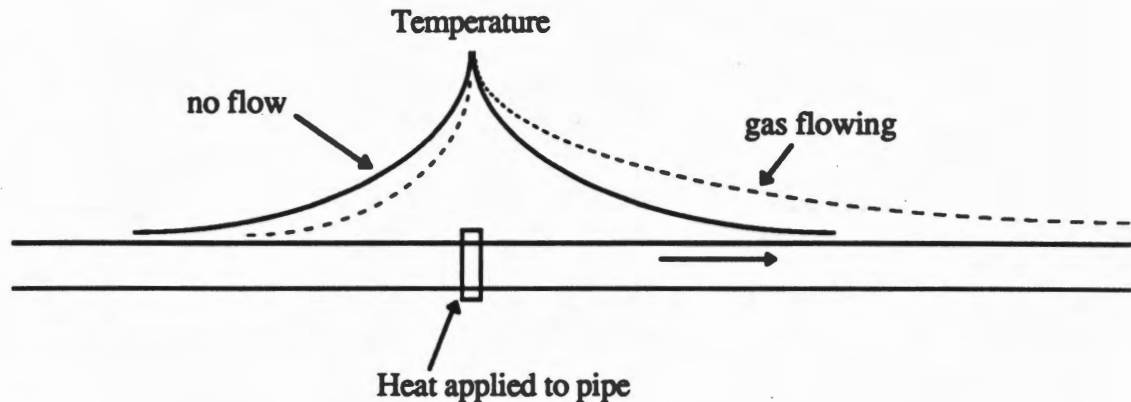


Figure 7.1. Temperature along Pipe for Gas Flow Measurement

2. Re-test the neutron attenuation technique for measuring  $^{235}\text{U}$  content in the 7.6-cm-diameter HEU pipe at Portsmouth after additional laboratory testing has been completed to ensure that the necessary electronic stability has been achieved. Also test the gamma-ray technique that uses the 186-keV gamma ray from  $^{235}\text{U}$ . The gamma-ray measurements could be conducted by Portsmouth or LANL, which have performed similar work in the past. Side-by-side comparison of both the neutron attenuation and gamma-ray methods could demonstrate the complementary aspects of the techniques and which are the most appropriate in the low gas pressure and elevated temperatures of the Portsmouth plant. The neutron attenuation tests will implement enhancements indicated by the stability tests, such as a highly stable data-acquisition system using readily available electronics and ensuring that the neutron counter is completely equilibrated for temperature and high voltage before beginning operations. Ideally, any demonstrations at Portsmouth should be done simultaneously with the neutron and gamma techniques (at least to the extent that the neutron source does not interfere with the gamma technique) to verify whether unusual results are due to plant operations or to problems in the measurement techniques.
3. If the temperature-profile technique fails to provide the necessary gas-flow results, then a pulsed-neutron generator could be used to replace the modulated  $^{252}\text{Cf}$  source in the delayed neutron counting system. The pulsed-neutron generator can provide a far greater signal and much lower background, which allows the determination of gas flow in less than an hour. The vicinity of the high neutron flux will need to be well shielded for radiation safety purposes, but otherwise the neutron counter would not require massive shielding. This option involves significant capital costs for the neutron generator and several months to implement. However, a high probability exists that this approach will provide the desired results on  $\text{UF}_6$  flow in the 20-cm (8-inch) LEU pipe.
4. If the pulsed-neutron generator technique is rejected for operational or cost reasons, then the modulated  $^{252}\text{Cf}$  technique could be implemented using all the improvements identified in this report. In particular, the pipe between the source and the detector needs to be shielded with polyethylene, and more polyethylene needs to be around the neutron source when it is in the background positions. Also, a faster multiscaling data-acquisition system needs to be

implemented, and any detector noise caused by operation of the motor drive must be eliminated. All these modifications be would be tested at PNNL before demonstrating the technology at Portsmouth. The preliminary tests might take several months to accomplish. These modifications should be adequate to accomplish the flow measurement task. However, it should be noted that the count rates will still be low, many hours of running will be required to obtain statistically significant data, and application would be limited to the 20-cm (8-in.) LEU pipe.

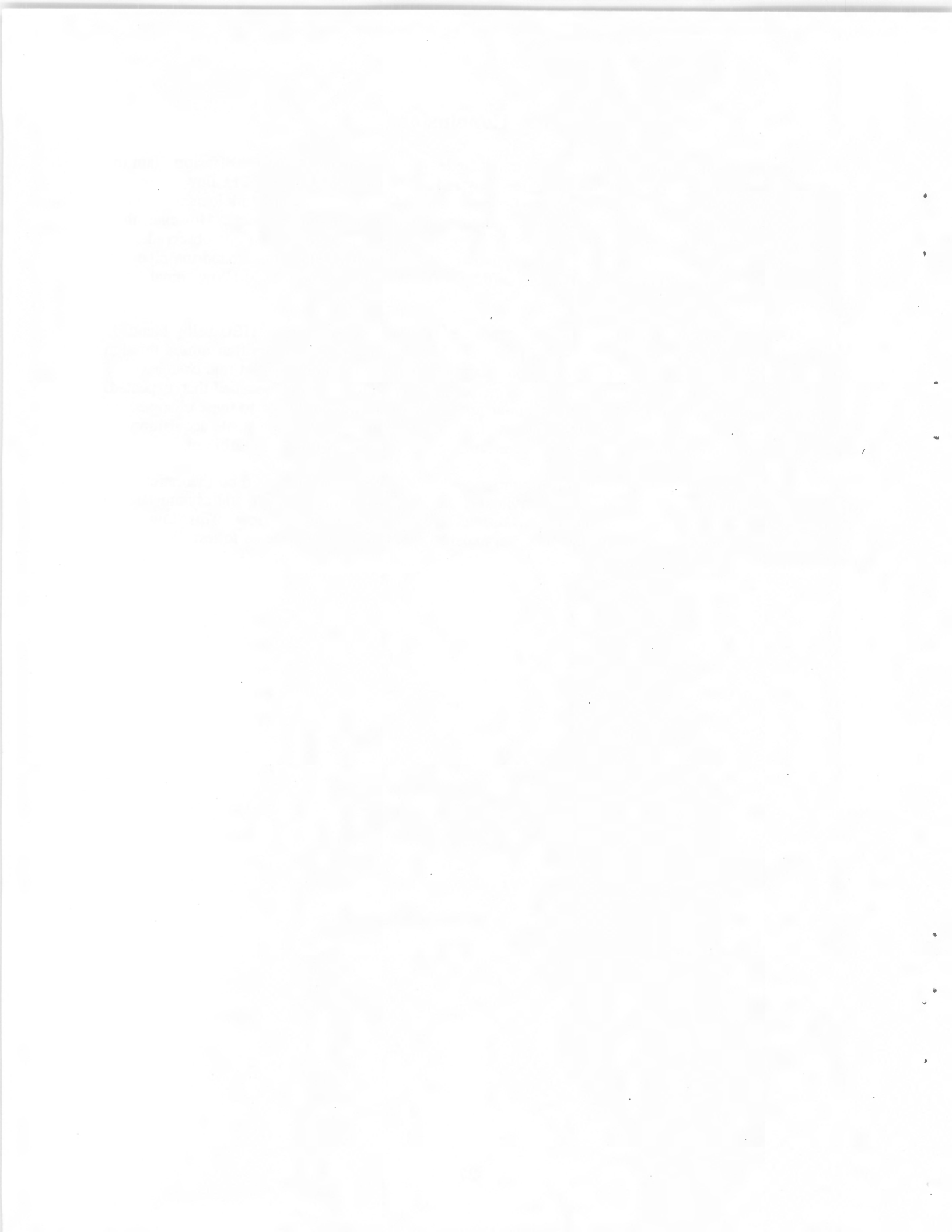


## 8.0 Conclusion

Two experiments using neutrons were conducted at the Portsmouth Gaseous Diffusion Plant to test non-destructive methods for measuring the flow and  $^{235}\text{U}$  content of  $\text{UF}_6$  gas. The flow measurement used a  $^{252}\text{Cf}$  source to produce fission in the  $^{235}\text{U}$ , and neutron detectors located downstream were designed to detect delayed neutrons emitted by the fission products. However, the neutron background from the source was too strong to allow the expected signal to be observed. Subsequent computer simulations have shown that added polyethylene shielding around the pipe carrying the gas and fission products should sufficiently reduce the background to allow signal detection.

A neutron attenuation experiment was conducted on the pipe carrying the HEU being blended to measure its  $^{235}\text{U}$  content. The presence of  $^{235}\text{U}$  reduces the number of neutrons transmitted through the gas because of thermal neutron absorption. Experimental results showed count rate changes when the HEU gas was removed. However, larger changes in count rate were obtained than expected. Apparently, electronic instabilities in the data-acquisition equipment contributed to these changes. The attenuation measurement will likely succeed in a longer-term test with more stable acquisition equipment and longer times provided for the electronics to reach temperature equilibrium.

A new, non-nuclear technique for measuring gas flow appears feasible based on theoretical calculations. In this technique, the pipe carrying the gas is heated at one location, and asymmetries in the temperature of the pipe upstream and downstream are produced by the gas flow. This flow measurement should work on any size pipe, but experiments need to be conducted to test the technique.



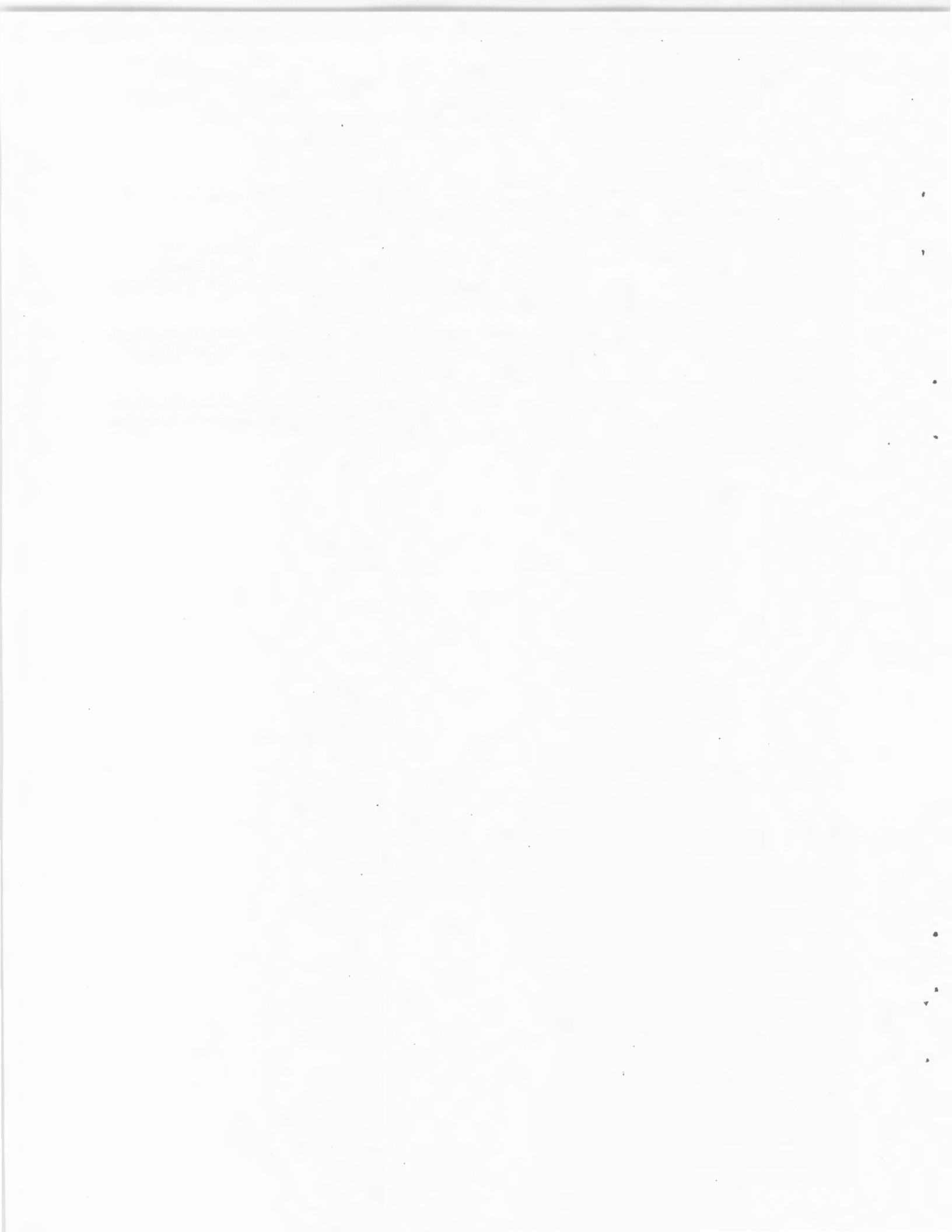
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