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Neutron Methods for Measuring ^{235}U Content in UF_6 Gas

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Summary

High-enrichment and low-enrichment UF_6 gas streams are blended in both the United States and Russia to reduce the inventory of highly enriched uranium. The product is suitable for fuel in nuclear power plants, but unsuitable for weapons production. Implementing a real-time process verification would help satisfy the international community that the blending process is truly reducing stocks of highly enriched material.

Computer simulations conducted at Pacific Northwest National Laboratory show that the ^{235}U content of UF_6 gas can be measured by monitoring the attenuation of thermal neutrons passing through pipes with a diameter as small as 2.5 cm and at UF_6 gas pressures as low as 5 mm Hg. The attenuation results from the preferential absorption of thermal neutrons by ^{235}U because of its high fission cross section. The long recoil ranges of the fission products in the low pressure gas do not degrade the measurement because the fission products do not contribute to the signal. At higher gas pressures (about 40 mm Hg) and larger pipe diameters (about 10 cm), an alternative method using a modulated neutron source and downstream detection of delayed neutrons will provide a measure of ^{235}U and flow speed of the UF_6 gas.

The Gaseous Diffusion Plant near Portsmouth, Ohio, where blending of UF_6 takes place, is a potential test location for non-intrusive methods to monitor blending. Testing both the neutron attenuation method and the modulated source method will be possible at that location. In addition, testing of a passive acoustic technique would show if noise generated by flowing UF_6 gas could be used to measure the flow velocity. It is necessary to test these methods to validate them before using them for specific applications.

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1.0 Introduction

In the United States and Russia, 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. A method to verify the blending of high- and low- enrichment uranium was developed at Pacific Northwest National Laboratory (PNNL)[a] for the U.S. Department of Energy, Office of Research and Development (NN-20).

In the United States, blending occurs at the U. S. Department of Energy's Portsmouth Gaseous Diffusion Plant located near Portsmouth, Ohio. In Russia, the blending takes place at Novouralsk. The United States is purchasing the blended product produced in Russia in a program to reduce the availability of enriched uranium that can be used for weapons production (Bieniawski and Dougherty 1995). Monitoring the ^{235}U mass flux of the input stream having the highly enriched uranium will provide confidence that high-enrichment uranium is being consumed in the blending process, and monitoring the output stream will provide an on-line measure of the ^{235}U in the mixed product. The Portsmouth plant is a potential test facility for non-destructive technology to monitor blending. In addition, monitoring the blending at Portsmouth can support International Atomic Energy Agency activities on controlling and reducing enriched uranium stockpiles.

Table 1 gives the parameters of importance for monitoring the blending process at Portsmouth and Russia. The main differences in the blending conditions that affect the monitoring at the two sites are the gas pressure and pipe diameter. The low pressure in small-diameter pipes at Portsmouth makes non-destructive monitoring difficult because the amount of UF_6 in the pipe is small, and the range of fission products is greater than the pipe diameter.

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 monitors the delayed fission neutrons emitted downstream in the flowing gas following the induced fission. This method was described in a previous report (Stromswold et al. 1996). A second method, described in this report, measures the attenuation of thermal neutrons crossing a pipe containing UF_6. The ^{235}U in the gas causes some of the neutrons to be absorbed, thus reducing the neutron flux reaching a detector located on the opposite side of a pipe from a source. This report also discusses a modulated-source method that uses neutron detection and an acoustic method for flow rate measurement.

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Table 1. Parameters for Blending UF_6

Location	Pipe	Enrich. (%)	Flow (m/s)	Pressure (mm Hg)	Temp (°C/ °F)	Pipe ID (cm)	Pipe OD (cm)
Portsmouth	Input (high ^{235}U) (a)	20 to 97	4.7	5	60 / 140	2.7	3.3
Portsmouth	Input (high ^{235}U) (a)	20 to 97	0.5	5	60 / 140	7.8	8.9
Portsmouth	Output	6	6.7	57	60 / 140	20	22
Russia	Input (high ^{235}U)	90	0.06	38	20 / 70	~ 9	~ 10
Russia	Input (low ^{235}U)	1.4	0.02	45	20 / 70	~ 9	~ 10
Russia	Output	4.4	1.0	30	20 / 70	~ 9	~ 10

(a) Portsmouth: input pipe for main blending location has inside diameter of 7.8 cm; input pipe of diameter 2.7 cm is at a backup blending location.

2.0 Fission Product Recoil Range

Monitoring methods based on downstream measurements of fission products require that the fission products stop in the UF_6 gas and be swept along by the gas stream. At the gas densities anticipated for the processes being monitored, stopping fission products in the gas may not always be possible.

Fission produces both light and heavy fission products that fly apart from each other at high energy (Figure 1). For low-pressure UF_6 gas, the range of these fission products can be much larger than the pipe diameter. Table 2 gives the estimated ranges of a light fission product (bromine) at two gas temperatures. As shown in the table, at a pressure of only 5 mm Hg (the pressure for Portsmouth's high enrichment input line), the fission product range is about 40 cm. In a 2.7- or 7.8-cm diameter pipe, most of the fission products will hit the wall of the pipe before they stop in the gas. Only the small fraction of particles traveling along the length of the pipe will stop in the UF_6 gas.

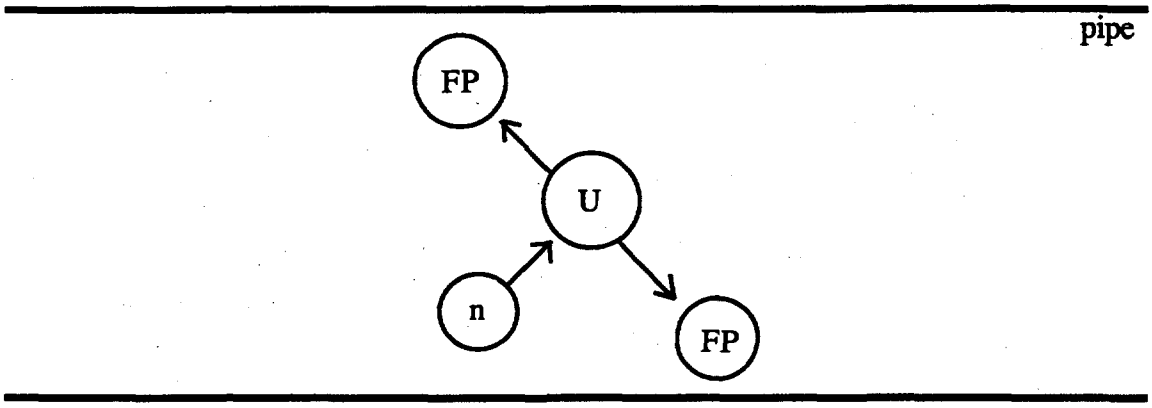

Figure 1. Recoil of Fission Products Inside Pipe Containing UF_6 Gas

The fission products that do stop in the gas travel significant distances upstream or downstream through the gas before stopping. This creates a broad interval for the starting location from which fission products subsequently drift downstream at the velocity of the gas flow. The broad starting interval makes gas-velocity measurement based on drift time difficult and reduces the signal variation available from a modulated neutron source.

Table 2. Range of Fission Products in UF_6 Gas

Gas Pressure (mm Hg)	Range of Fission Products (cm)	
	Temp = 20°C	Temp = 60°C
5	38	44
30	6	7
40	5	5
60	3	4

3.0 Thermal Neutron Attenuation Method

Induced fission can provide a measure of the ^{235}U content even at low UF_6 gas pressure. However, the neutron signal of most use at low gas pressure is the attenuation of thermal neutrons passing though a pipe containing UF_6 gas. The attenuation results from absorption of thermal neutrons when fission occurs in the ^{235}U. The long travel range of the fission products does not hinder the attenuation measurement because fission products do not contribute to the signal.

Figure 2 shows a cross section of the equipment configuration for the proposed method of thermal neutron attenuation. A ^{252}Cf source is located on one side of a pipe containing UF_6, and a ^{3}He neutron detector is on the other side of the pipe. Polyethylene surrounding the entire system out to a distance of about 20 cm moderates the energetic neutrons from the source, thereby producing thermal neutrons that scatter back and forth through the pipe. Cadmium located on all sides of the detector, except toward the pipe, absorbs thermal neutrons that have not passed through the UF_6. Some of the thermal neutrons are absorbed by ^{235}U as induced fission occurs. Thus the count rate in the detector decreases with increasing ^{235}U content.

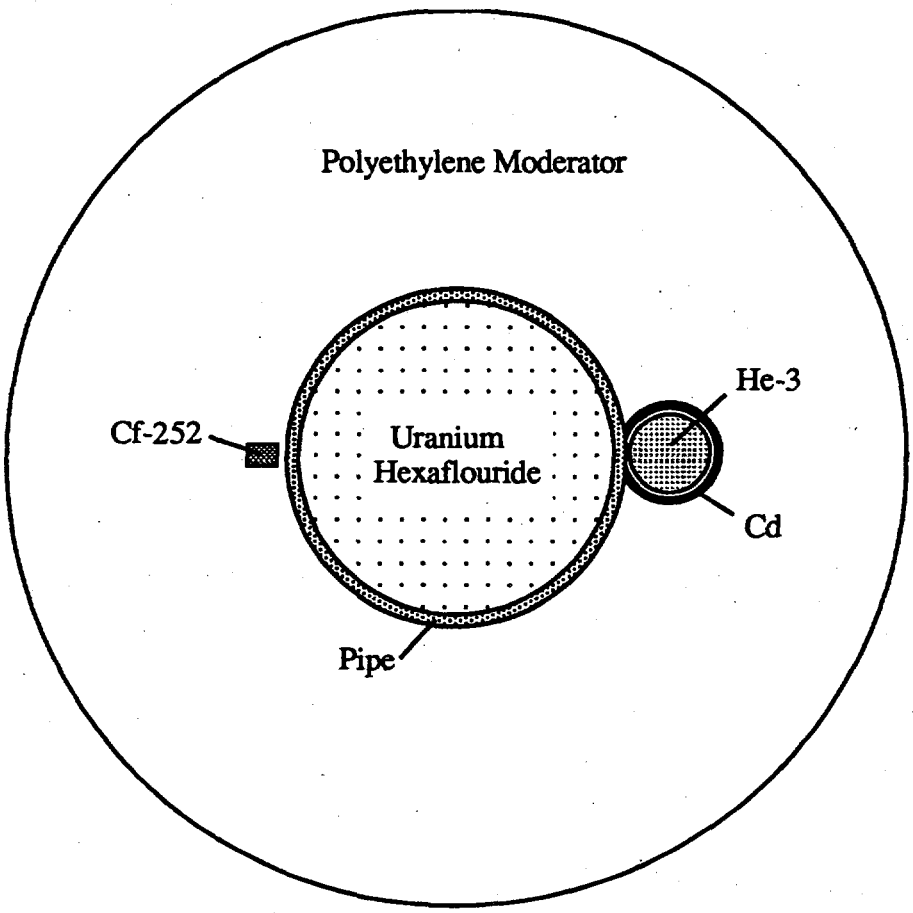

Figure 2. Configuration for Measuring Thermal Neutron Attenuation

3.1 Computer Simulations

Computer simulations using the code Monte Carlo Neutron Photon (MCNP version 4A; Briesmeister 1993) guide the design of the detection equipment and give anticipated count rates and measurement sensitivities. For the MCNP simulations, the neutron detector is a single ^{3}He tube 36 cm long with a pressure of 4 atmospheres. The cadmium is 1-mm thick, and its opening at the pipe is modeled with various widths to determine optimum collimation. The simulated steel pipes have sizes and wall thicknesses appropriate for the Portsmouth plant. The polyethylene around the pipe has a radial thickness of 8.5 cm and a length of about 50 cm. The simulated UF_6 gas has a pressure of 5 mm Hg for the 2.5- and 7.6-cm diameter pipes and 50 mm Hg for the 20-cm diameter pipe. The ^{235}U enrichment is 0% or 100% in all simulations.

For a given physical configuration of neutron source, pipe, and detector, the measurement sensitivity is characterized by the detected count rate and by the change in the count rate when ^{235}U is present. The goal of the MCNP simulations is to maximize the count rate in the neutron detector (detection efficiency) while simultaneously providing the greatest fractional change in count rate (attenuation factor) as a function of the ^{235}U content of the gas in the pipe. The simulations show the following results:

- *Neutron source energy and position:* The MCNP simulations vary both the neutron source energy (about 2 MeV to simulate a ^{252}Cf source and 0.3 MeV to simulate an AmLi source) and the source position with respect to the pipe. Results show that the detection efficiency is about twice as large using the low-energy source as for the high-energy source. However, the attenuation factor remains the same for both sources. The ^{252}Cf source is selected for subsequent calculations and for experimental implementation because AmLi sources are limited in neutron output. The MCNP simulations also show that varying the source position within a few centimeters of the pipe has little effect on the detection efficiency and attenuation factor.

- *Collimation:* The collimation of the detected thermal neutrons is affected by the size of the opening in the cadmium around the neutron detector. The MCNP simulations show that reducing the opening to produce greater collimation between the source and the detector both decreases the detection efficiency and the attenuation factor. The widest opening that still requires accepted neutrons to pass through the gas is preferable.

- *Multiple detectors:* For pipes with diameters larger than 2.5 cm, multiple 2.5-cm diameter detectors are simulated side-by-side near the pipe. The additional detectors significantly increase the detection efficiency, but with little enhancement to the attenuation factor. Multiple detectors may thus shorten the counting time, provided the electronics can handle the increased count rate. Although the larger diameter pipes have room for additional detectors adjacent to them, additional detectors are not necessary because the desired attenuation of thermal neutrons passing through the UF_6 in these pipes is much larger than in the small pipes.

Table 3 shows the MCNP simulation results for a single detector located opposite a ^{252}Cf source for pipes of the three diameters found at Portsmouth. As shown in the table, the detection efficiency (counts/second per neutron/second from source) is largest (1.32%) for the 2.5-cm pipe and smallest (0.318%) for the 20-cm pipe. In contrast, the attenuation factor (change in counts when enrichment changes from 0 to 100%) is largest (1.4%) for the 20-cm pipe. Also shown in the table is the statistical uncertainty in the calculated ^{235}U content, assuming 100% enrichment. This uncertainty ranges from 19% for the 2.5-cm pipe to 0.6% for the 20-cm pipe. The calculated uncertainty assumes a 10^7 n/s neutron source and a counting time of 10^4 s (2.8 h). The uncertainty arises from subtracting two large numbers (the counts with no ^{235}U minus the counts with 100% ^{235}U) as follows:

N_1 = counts with no ^{235}U = (source strength)(counting time)(detection efficiency)

= (10^7 n/s)(10^4 s)(0.0132) [for 2.5-cm pipe]

= 1.3200 x 10^9 counts

N_2 = counts with ^{235}U = (source strength)(counting time)(detection efficiency)(1- atten.)

= (10^7 n/s)(10^4 s)(0.0132)(1- 0.0002) [for 2.5-cm pipe]

= 1.3197 x 10^9 counts

Uncertainty (1s) = [Sqrt (N_1 + N_2)] / (N_1 - N_2)

= 0.19 [for 2.5-cm pipe]

Although the uncertainties listed in Table 3 are rather large for the 5-mm gas pressure conditions, it should be emphasized that measurements of ^{235}U content appear to be impossible by other non-invasive techniques. Even with the large uncertainties, the attenuation measurements provide the necessary assurance that the process blending conditions are close to the stated conditions.

The MCNP calculation results shown in Table 3 assume 100% enrichment of the UF_6 gas. For the 2.5- and 7.6-cm diameter pipes at Portsmouth, this enrichment approximates the actual condition. However, in the 20-cm diameter pipe the actual enrichment is only about 6%. When this lower enrichment is included in the uncertainty calculation, the resulting uncertainty for the ^{235}U content is about 9%, rather than 0.6%.

Table 3. MCNP Results for Attenuation of Thermal Neutrons in 100%-Enriched UF_6 Gas

Pipe Diam. (cm)	Pressure (mm Hg)	Detection Efficiency (c/s/n/s; %)	Attenuation (%)	Uncertainty in Calc. ^{235}U (%)
2.5	5	1.32	0.02	19
7.6	5	0.59	0.05	12
20	50	0.32	1.4	0.6

The thermal neutron attenuation method does not provide a means for measuring the flow velocity of the UF_6 gas. A separate measurement, perhaps non-nuclear, would be required for the velocity. Monitoring the flowing gas acoustically to determine velocity is discussed in a separate section of this report.

3.2 Anticipated Testing at Portsmouth

Equipment to perform tests of the neutron attenuation method has been assembled at PNNL. For the anticipated testing at Portsmouth, the polyethylene is positioned around the 7.8-cm and 20–cm pipes in a configuration similar to that of Figure 2, except that the shape of the polyethylene moderator is rectangular rather than a cylindrical for ease of construction. No test of the 2.7-cm pipe is anticipated during the initial tests because that diameter pipe is not yet being used at Portsmouth,

where both the main blending station and the secondary station use 7.8-cm diameter pipes for the high enrichment UF_6 flow.

The polyethylene moderator, ^{252}Cf source (emitting 10^7 n/s) and detector are located near the blending station. The temperature at this location is 60°C (140°F). Supporting electronics, except for possibly the preamplifier for the ^3He detector, are located outside the cell where the temperature is cooler. Figure 3 is a schematic of the equipment to be used. Figure 4 is a photograph of the polyethylene moderator and detector to be used with the 7.6-cm diameter pipe. A similar moderator exists for the 20-cm diameter pipe.

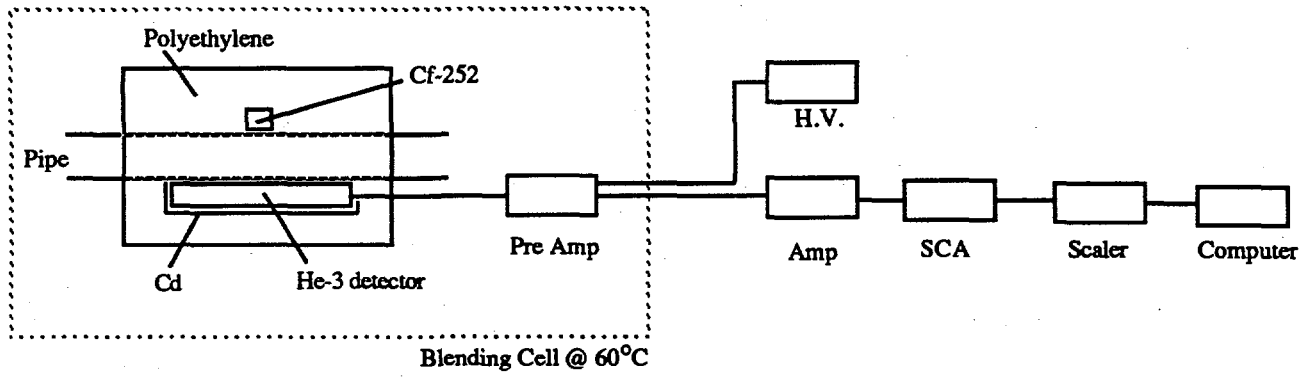

Figure 3. Equipment for Neutron Attenuation Test at Portsmouth

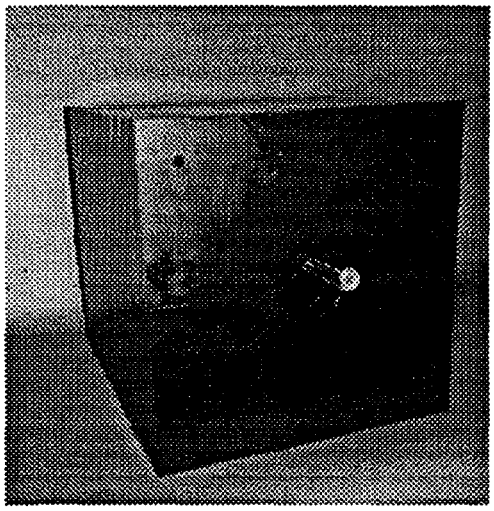

Figure 4. Polyethylene Moderator for Thermal Neutron Attenuation Measurements through 7.6–cm Diameter Pipe at Portsmouth

The tests at Portsmouth will give experimental data on the attenuation of neutrons passing through the pipe. The attenuation is due to fissions induced in the UF_6 gas and in any UF_6 on the walls of the pipe. To separate the two effects, the UF_6 gas needs to be removed from the pipe for part of the experiment to obtain "no-gas" background data. During the no-gas experiment, the pipe can be evacuated, or it can contain nitrogen or some other non-fissioning gas.

The neutron background at the Portsmouth facility should not pose any problem for the attenuation measurement. Indeed, the ^{252}Cf source that provides the source neutrons at a rate of 10^7 n/s creates a much greater neutron flux in the measurement region than does any anticipated neutron background. In analyzing data for the attentuation technique, two large counts are subtracted to obtain the attenuation caused by induced fission of ^{235}U. As shown previously in Section 3.1 for the MCNP simulations, the statistical uncertainty in the attenuation measurement results from subtracting two large counts corresponding to the counts obtained from the ^{252}Cf source with and without ^{235}U present in the pipe. Any neutron background at the Portsmouth facility is likely to produce counts in the detector that are much smaller than those from the ^{252}Cf source. Consequently, the effect of the neutron background on the determination of ^{235}U content and its statistical uncertainty is negligible.

4.0 Modulated Neutron Source Method

A modulated neutron source produces time-varying induced fissions in the UF_6 gas. The fissions create fission 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}U content of the gas and on the gas-flow velocity. The amplitude of the signal depends on the ^{235}U content of the gas, and the phase shift of the signal, compared to the source modulation, gives the flow velocity. J. T. Mihalczo at Oak Ridge National Laboratory proposed a similar technique that uses a modulated neutron source, but detects delayed gamma rays when sufficient UF_6 gas is present (Mihalczo et al. 1996). Both the delayed gamma-ray and neutron detection methods are distinct technologies with their individual benefits and disadvantages. Testing under realistic operating conditions at a uranium blending plant will provide the best evaluation of the two techniques.

At low gas pressures of 5 mm Hg and pipe diameters of 2.7 and 7.8 cm, the modulated technique is unlikely to work for either neutrons or gamma rays because the signal is too small. In addition, the long recoil distances of the fission products in low-pressure gas imbeds most of the fission products in the pipe walls or broadens the starting location from which fission products subsequently drift downstream at the velocity of the gas flow. The broad starting interval makes gas velocity measurement more difficult and reduces the signal variation available from a modulated neutron source.

The modulation method will likely produce a measurable signal for the 20-cm pipe at Portsmouth containing UF_6 at 57 mm Hg because of this pipe's larger diameter and higher gas pressure. Experiments are necessary, however, to prove the performance. Figure 5 shows the anticipated equipment for a modulated source and downstream measurement of delayed neutrons. Source modulation is achieved by a pneumatic piston that rapidly moves a ^{252}Cf source between two different positions: one position near the pipe and the other away from the pipe (a distance of about 20 cm). When the source is located near the pipe, more fissions are induced in the ^{235}U than when the source is away from the pipe. The downstream neutron detector (Figure 6) is an array of 36 ^{3}He tubes 36 cm long with diameters of 2.5 cm. The tubes are arranged around the pipe in two concentric circles and embedded in polyethylene. The separation between the source and the detector array is about 3 m. For a flow velocity of 6.7 m/s, the transit time of the gas between the source and detector will be about 0.4 s. The source anticipated modulation period is 5 to 10 s with about 0.1 s being required for the pneumatic piston to move the ^{252}Cf source.

The MCNP calculations presented in a previous report (Stromswold et al. 1996) give the number of expected induced fissions in the UF_6 for the Russian blending conditions per source neutron emitted from a ^{252}Cf source. Adjusting those results to the gas pressure, temperature, and pipe diameter for Portsmouth gives an estimate of the induced fission rate for a source located adjacent to the pipe. For the 20-cm pipe at Portsmouth containing 6% enriched UF_6, the induced fission rate in the gas is about 2×10^3 fissions/s near a ^{252}Cf source emitting 10^7 n/s . Neutron detectors located 3 m downstream would have a count rate of about 0.2 n/s from these fissions. This count rate should be discernable from background, which is expected to be about 1.5 c/s when the neutron source is moved to its position away from the pipe. Modulation greatly improves the ability to distinguish signal from background. Standard fourier analysis of the time-varying signal gives a powerful tool for processing data.

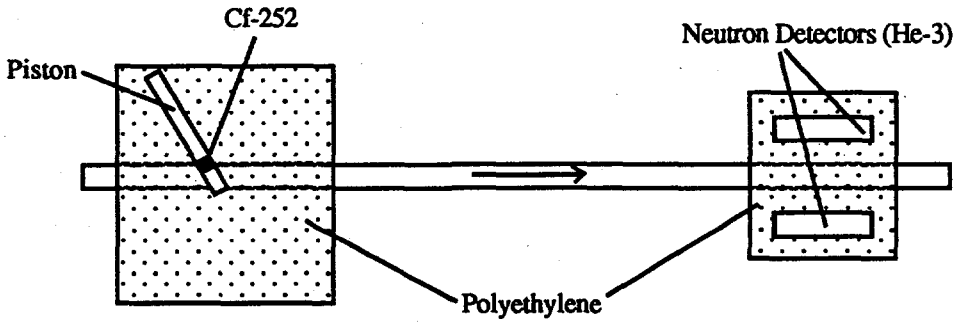

Figure 5. Modulated Source with Delayed Neutron Detection

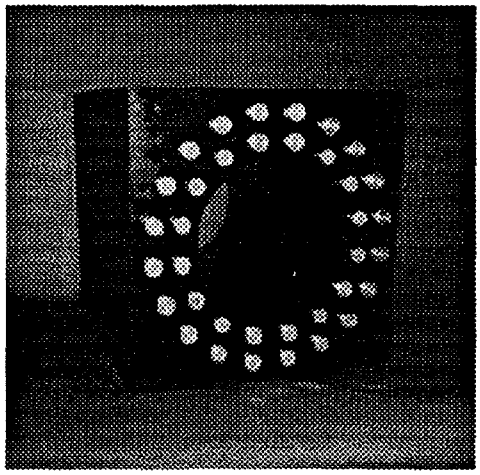

Figure 6. Neutron Detectors in Polyethylene Moderator for Use with Modulated Source on 20–cm Diameter Pipe at Portsmouth

5.0 Acoustic Emission Monitoring for Flow

Ultrasonic techniques are routinely used in industry to measure the rate of fluid flow continuously. Several commercial vendors, including Panametrics and Scientific Engineering Instruments, offer off-the-shelf equipment for applications involving liquids in piping and gas effluents in stacks. The common techniques entail precise time-of-flight (TOF) measurements or Doppler-shifted frequency analyses. However, no commercially available equipment is suitable for measuring the rate of gas flow in a pipe from the pipe's outer surface noninvasively.

The present technical limitation is the inherent signal loss associated with injecting high-frequency acoustics through a metal wall into a gas. The technical approaches presently applied in commercial equipment employ acoustic frequencies that are conducive to accurate TOF measurements, but are not suitably applied for sampling gases through metal piping. Tests at PNNL using a flow loop to simulate UF_6 gas flow used low frequency (5 to 100 kHz), high-energy acoustics for minimizing the inherent insertion losses from the active source and advanced signal processing techniques for the required TOF measurement accuracy. The test results show the difficulty of using this technique to measure flow velocity at low gas pressures in thick-walled pipes. The technical limitation of the technique is the extraction of gas-flow information from the innocuous background noise generated in thick-walled, nominal 6-mm pipe. The approach was demonstrated successfully only on thin-walled, nominal 1.5 mm, laboratory material. It is clear that for gas pressure as low as 100 mm Hg and thick-walled pipes, this acoustic method for flow velocity does not work.

A second ultrasonic technique for a rate of fluid flow measurement is based on sampling natural acoustic emissions associated with fluid flow passively. Similar approaches have been applied successfully to the detection of leaks in gas and liquid transport piping and in the monitoring of product flow in food-processing facilities. The proposed approach is strictly empirical, requiring a calibration procedure using known flows. The specific limitations have not yet been explored, but the approach is based physically on the existence of some irreversible loss mechanism in the fluid, such as found in turbulent flow. Cursory estimations of the Reynolds numbers for the Portsmouth lines suggest that prospects of such loss mechanisms in straight pipe sections far away from orifices are marginal. However, acoustic emission sensors could be placed near pipe constrictions, bends, or installed orifices where turbulent flow is more likely.

Experiments with the passive acoustic technique at Portsmouth will show if this method is practical. Sensors could be attached to the pipe at several locations, preferably at bends or constrictions in the pipe, to obtain passive acoustic signatures over a wide frequency spectrum. Comparing the acoustic signatures from different locations along the pipe should allow background noise from pumps and other machinery to be subtracted, perhaps leaving a portion of the frequency spectrum that can be correlated with gas flow. If the acoustic noise created by the flow of UF_6 gas presents a method for measuring flow rate, that technique can be coupled with the thermal neutron attenuation method for determining ^{235}U content.

6.0 Conclusions

Computer simulations show that the attenuation of thermal neutrons passing through UF_6 gas in a pipe will give a method for measuring ^{235}U content, even down to pressures of 5 mm of Hg. The attenuation is proportional to ^{235}U content because thermal neutrons are removed by induced fission. The large fission product range and the collision of the fission products with pipe walls does not present a problem for this measurement. Although the signal is small and the resulting uncertainties can be rather large in some cases (e.g., 12% for 7.6-cm-diameter pipe), the non-intrusive techniques described still provide a level of certainty that blending is occurring.

At higher gas pressures of 40 to 60 mm of Hg, the range of the fission products is sufficiently short so that most of the fission products stop in the UF_6 gas and then drift with the gas flow. The delayed neutrons emitted from the fission products can be measured downstream to determine ^{235}U content. Modulating the neutron flux that causes the induced fission gives both an improved measure of the signal for determining the ^{235}U content and a way to measure flow velocity.

The gaseous diffusion plant at Portsmouth, Ohio, offers a location for testing the attenuation and modulated neutron methods, plus the passive acoustic method, on actual UF_6 gas flows. Experiments should be conducted to demonstrate the feasibility of these methods under plant operating conditions.

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