MONITORING THE FLOW OF FISSILE LIQUIDS

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ABSTRACT

Monitoring the flow of fissile liquids between tanks or facilities can be accomplished by inducing fission in the fissile material with a modulated neutron source and detecting delayed gamma rays downstream. The modulation of the neutron source produces waves of activation whose delay can be detected in gamma sensitive detectors downstream to obtain the flow velocity. The detector count rate can be related to the fissile mass in the liquid. This system can be attached to the outside of pipes and utilizes a polyethylene moderator, a neutron absorbing material that can be moved in and out between the pipe and the surrounding moderator, and a detector of delayed gamma rays downstream. This system is similar in principle to systems that have been used to monitor the flow of UF_6 gas in the blend-down of HEU to LEU in Russia. The high density of the fissile material in the liquid (grams to 100's of grams per liter) of solution compared to the very low density of UF_6 gas results in an implementation of this method that uses much smaller sources and much less moderator since the fissile liquids usually contain moderator.

This paper describes the Monte Carlo neutron and gamma transport calculation used to design the system, the system design, and expected performance for some typical liquids and flows. This type of system has applications for nuclear safeguards. It also has application for criticality safety since flow into non-geometrically safe tanks can and has resulted in criticality accidents. Although these simulations were for uranyl nitrate, the methodology could be applied to Pu solutions with slightly longer measuring times (or larger source sizes).

Keywords: Uranium Flow Correlation Shutter

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INTRODUCTION

Monitoring the flow of fissile liquids between tanks or facilities can be accomplished by inducing fission in the fissile material with a modulated neutron source and detecting delayed gamma rays downstream. The modulation of the neutron source produces waves of activation whose delay can be detected in gamma sensitive detectors downstream to obtain the flow velocity.

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METHOD

Figure 1 is a conceptual sketch of the measurement apparatus. The method is simple: a neutron shutter periodically opens, allowing ^{252}Cf neutrons to pass into the pipe and induce fissions in the U traveling down the pipe. These fissions create daughter products that later emit gamma rays in front of the downstream detector. Since the ^{252}Cf shutter opens briefly, there is a brief rise of gamma ray emissions at the detector when the irradiated flow passes the detector. The time between the shutter opening and the detector's count increase is the flow transport time. The flow rate is calculated from the measured transport time. The system uses gamma rays because there are many more delayed gammas than delayed neutrons.

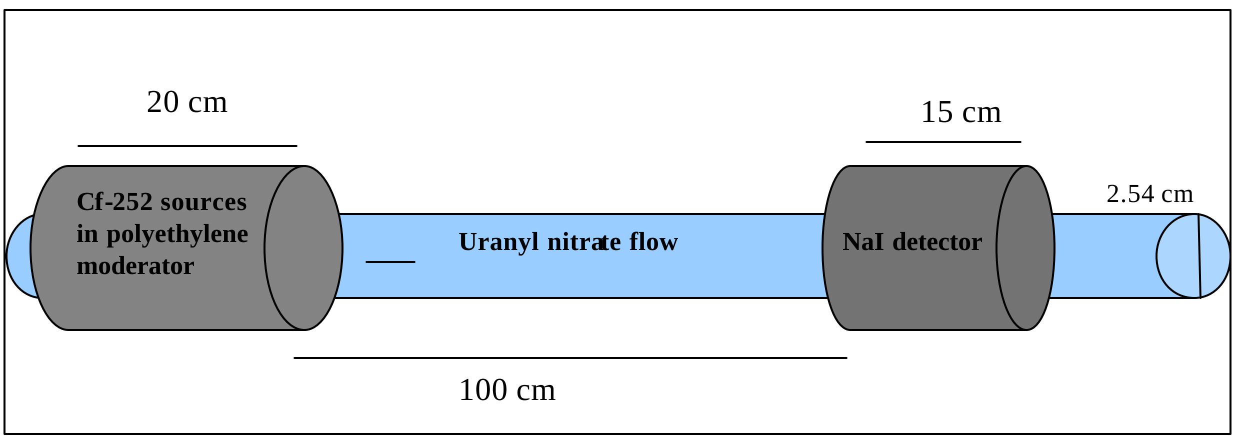

Figure 1 Sketch of the measurement apparatus showing the major elements.

In this configuration the flow pipe is schedule 40 stainless steel pipe with 1" inside diameter. The source and detector are separated by 39.37" (1 meter). The pipe contains a uranyl nitrate solution with 93% enrichment.

The ^{252}Cf source is positioned in polyurethane both to shield it and to moderate the neutron energies to maximize pipe fissions. The ^{252}Cf source is in a polyethylene collar around the pipe with 4.6 cm ID and 14.8 cm OD. The optimum source depth in the polyethylene was determined by simulations as show in Figure 2. Thus a 1 μg ^{252}Cf source induces 18200 fissions/sec in the pipe.

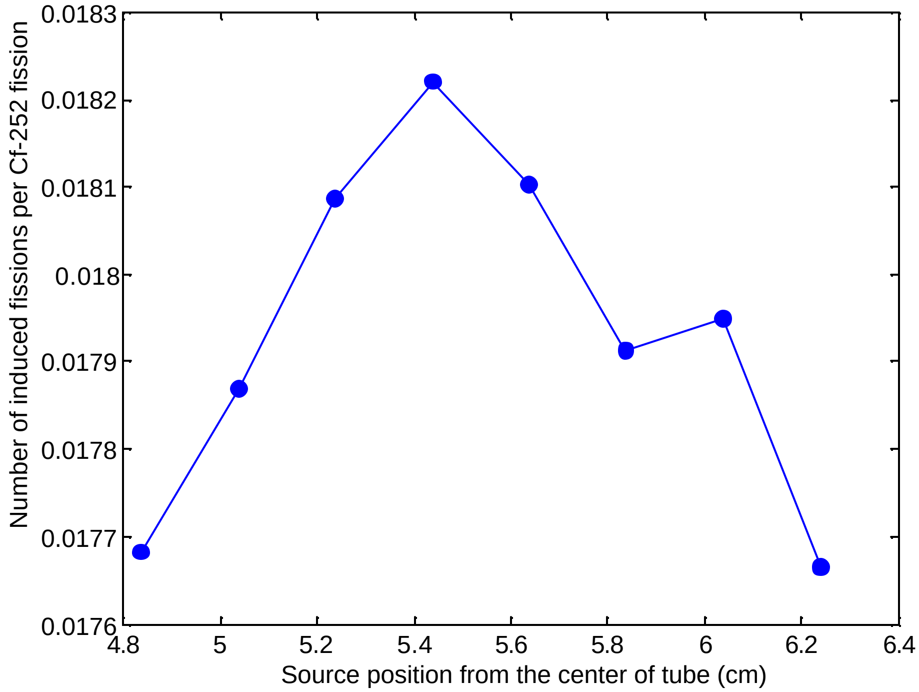

Figure 2 Optimization of source position in polyethylene moderator. Source position is measured from the center of the pipe.

The neutron shutter contains Li^6 and reduces the pipe fission rate by 83% when it closes.

This configuration uses a NaI detector in the shape of a collar around the pipe, with 5.7 cm ID and 15.8 cm OD. NaI is used because it is sensitive to gamma rays but not to neutrons. In this configuration the detector efficiency for pipe gamma ray radiation is 30%. There is a 5 cm lead shield on the source-side of the detector to block the source's gamma rays. With a 10^6 ^{252}Cf source the direct radiation to the detector is 2301 cps with the neutron shutter open and 2231 cps with the neutron shutter closed.

The total weight of this configuration is 196 lbs (88.5 kg).

The concept comes from a U gas-flow monitor that has operated in Russia for several years [1]. This paper describes a U liquid-flow monitor. Since the U is much more dense in the liquid than the gas, this is a much easier application, but the experience gained with the gas-flow monitor is reflected in this design. However this configuration presented here has not be optimized and some useful techniques used in the gas flow monitoring system are not employed here.

The neutron shutter opens periodically to irradiate the flow. The shutter's optimum movement schedule is determined by two factors. First, it opens long enough to produce an irradiated slug of U solution in the entire length of pipe in front of the detectors. This produces the maximum gamma ray count in the detectors. Second, it stays closed long enough that it is not open twice during the flow's transport between source and detectors. This allows the system to employ a simple method to determine the flow transport time between source and detector. Multiple openings of the shutter would produce multiple gamma count peaks during the flow transport time. Both of these factors depend on the range of flow rates to be measured. (In this paper, flows between 1 and 12 gpm were studied.)

Radiation reaches the detector through 3 paths:

- Immediate, direct radiation from the source through the shielding around the source and the detector,
- Immediate fission radiation from the pipe adjacent to the source directly to the detector,
- Delayed gamma radiation from daughter products in the pipe to the detector (the radiation that the measurement depends upon).

The immediate radiation is a nuisance since it does not depend on the flow transport delay. Furthermore, it varies with the neutron shutter position so it is also time-dependent. However it can be subtracted from the signal if need be. This was done for the gas flow monitor but is not necessary for this liquid flow application due to the strong delayed gamma component.

If one observation of the shutter movement does not yield a clear indication of the flow transport time, multiple observations can be summed as needed. This is necessary when very weak radiation sources are used. Since the correct flow rate can be obtained (eventually) by summing enough observations, the performance of the system is expressed by the number of observations required to get an accurate flow measurement.

SIMULATION RESULTS

Simulations were performed for a variety of flow rates and ^{252}Cf source sizes. Flow rate was varied between 3.8 to 45.4 liters per minute (1 and 12 gpm), so that the flow travel time lay between 2.7 and 32 seconds. The source strength was varied between 0.03×10^6 and 1.00×10^6 fissions per second.

The system measures the number of detector counts for every 0.1 second interval during flow. Figure 3 illustrates the detector count signal. For this illustrative case the source was 1 µg of ^{252}Cf undergoing 10^6 fissions per second. The shutter opened for 2.6 seconds at a time, and the liquid flowed between source and detector in 6.4 seconds.

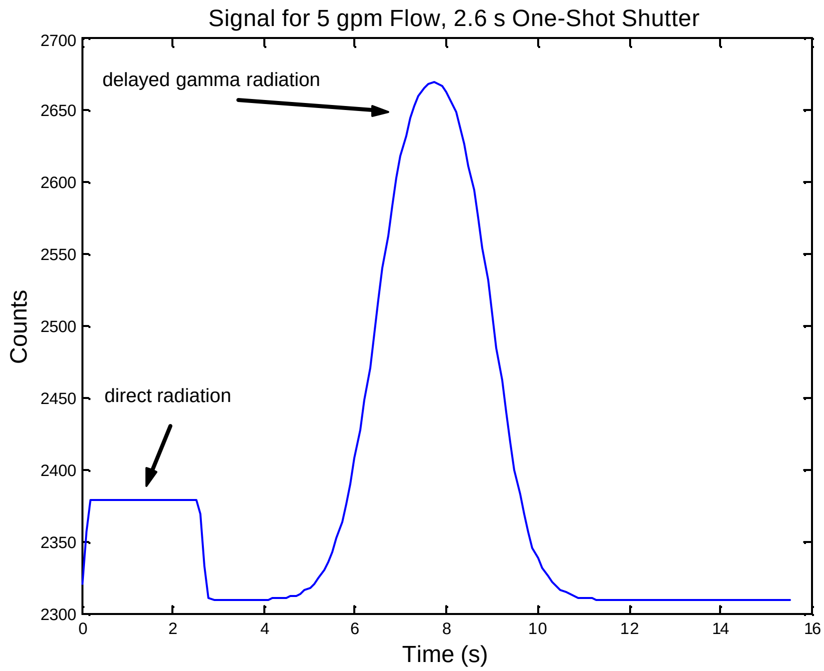

Figure 3 Example of the detector signal showing the radiation directly from the source and the radiation from the delayed gamma rays in the pipe's Uranium solution flow.

The flow transport time is found by calculating the cross correlation $C(t)$ between the shutter movement $x_s(t)$ and the detector count time series $x_d(t)$, over an observation period *T*, as follows:

$$C_{ij}(t) = \frac{1}{T-t}\sum_{t=0}^{T-1-t} x_s(t)x_d(t+t)$$

The cross correlation is calculated as a function of the time lag *t* between a shutter movement and the detector signal; the correlation's maximum occurs when *t* is the flow transport time. The correlation's benefit is its ability to identify the flow time when the detector signal is noisy. For example, Figure 4 shows a noisy detector signal due to the use of a much smaller source producing 10^4 fissions per second. Figure 5 shows the correlation for this measurement, which peaks at the correct flow time.

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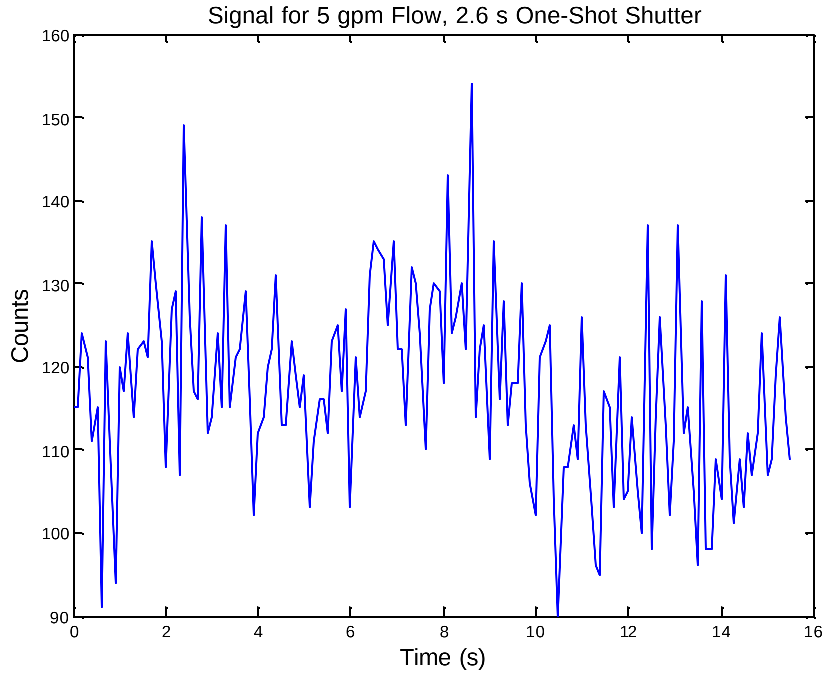

Figure 4 Detector signal using a very low source strength.

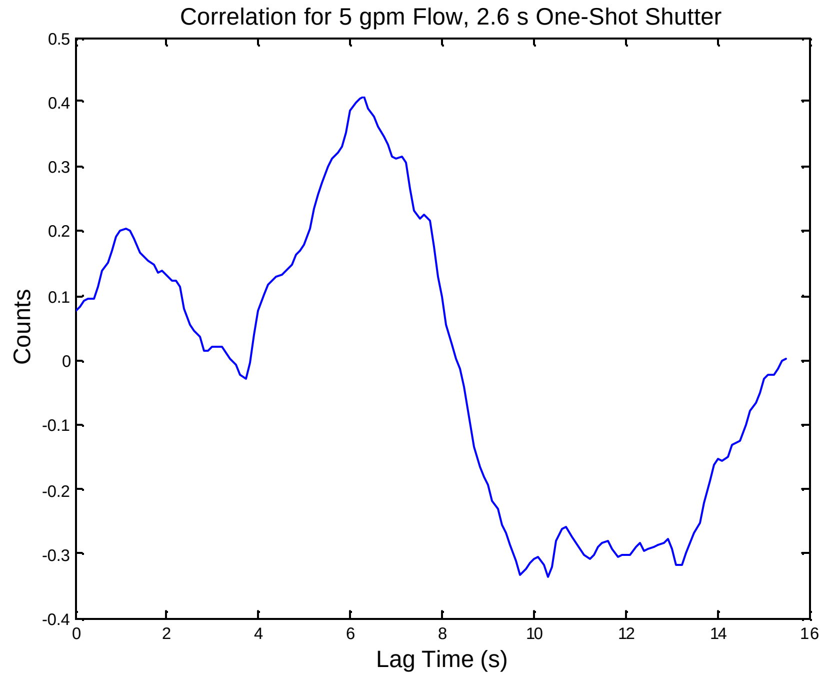

Figure 5 Correlation of the noisy detector signal with the neutron shutter position using a very low source strength. The correlation maximum occurs at the flow transport time.

A study examined the limits of performance for this configuration of the system. The range of flow rates required an observation time of 77 seconds per source pulse and a shutter opening time of 12.9 seconds. The performance was tested by randomizing the detector counts for 400 trials, requiring that 95% of the trials measure the correct flow, determining how many observations must be summed to achieve this. (A measurement was deemed to be correct if it was within 10% of the actual flow rate.) Table 1 shows the total observation time (seconds) required for each combination of flow rate and source size to achieve 95% reliability.

		flow rates, gpm					
		1	4	6	8	10	12
source size, millions of fissions per second	1.00	77	77	155	77	77	155
	0.70	77	155	155	77	77	232
	0.50	77	77	232	155	155	232
	0.40	77	155	232	155	155	310
	0.30	77	155	310	155	232	387
	0.25	77	232	387	232	232	464
	0.20	77	310	464	232	310	542
	0.15	77	387	542	310	387	619
	0.10	77	697	697	542	619	929
	0.05	77	929	1316	929	1161	2090
	0.03	155	1316	2864	1625	2090	3715

Table 1 The limits of accuracy of the flow measurement were explored using low source strengths. The total observation time (seconds) required for 95% reliability is shown for each combination of flow rate and source strength.

The accuracy of this measurement could be extended to higher flow rates and lower source sizes by employing additional techniques used in the gas flow monitoring application. First, the direct radiation from source to detector could be subtracted from the detector signal before calculating the correlation. This would remove the zero lag component of the signal and correlation, avoiding any confusion between that and the true flow transport time. Second, the shutter could move much more frequently in order to create more variation in the detector signal. Opening the shutter more often will produce more pipe fissions and better averaging for the correlation measurement. Third, more distance between the source and detector would increase the flow transport time and allow for a more precise measurement at the higher flow rates.

CONCLUSIONS

A flow monitoring technique used for U gas flows for several years was simulated for liquid U flows. The liquid U flow application was found to be much simpler due to the higher density of U in the liquid. A simpler measurement technique using small ^{252}Cf sources was found to be successful for a wide range of liquid flow rates. The more complex techniques employed for the gas flow monitor could be employed to cover a wider range of flow rates or allow smaller sources, if necessary. Also, no attempt has been made to optimize this design's geometry or detector system so more improvements should be possible. For example, more separation between the source and the detector would improve the performance for higher flow rates; a BGO detector might be a better choice due to its higher density.

This type of system has applications for nuclear safeguards. It also has application for criticality safety since flow into non-geometrically safe tanks can and has resulted in criticality accidents.

REFERENCES

1. “Methodology for Interpretation of Fissile Mass Flow Measurements,” José March-Leuba, J. K. Mattingly, J. A. Mullens, T. E. Valentine, J. T. Mihalczo, R. B. Perez, 38th Annual Meeting of the Institute of Nuclear Materials Management, Phoenix, Arizona, July 20, 1997.

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