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ABSTRACT:

As part of an ongoing effort at Lawrence Livermore National Laboratory (LLNL) to enhance analytical models that simulate enrichment and conversion facilities, efforts are underway to develop routines to estimate the total gamma-ray flux and that of specific lines around process piping containing UF_6. The intent of the simulation modeling effort is to aid in the identification of possible areas where material diversion could occur, as input to an overall safeguards strategy. The operation of an enrichment facility for the production of low enriched uranium (LEU) presents certain proliferation concerns, including both the possibility of diversion of LEU and the potential for producing material enriched to higher-than-declared, weapons-usable levels. Safeguards applied by the International Atomic Energy Agency (IAEA) are designed to provide assurance against diversion or misuse. Among the measures being considered for use is the measurement of radiation fields at various locations in the cascade hall. Our prior efforts in this area have focused on developing a model to predict neutron fields and how they would change during diversion of misuse. The neutron models indicated that while neutron detection useful in monitoring feed and product containers, it was not useful for monitoring process lines. Our current effort is aimed at developing algorithms that provide estimates of the gamma radiation field outside any process line for the purpose of determining the most effective locations for placing in-plant gamma-monitoring equipment. These algorithms could also be modified to provide both dose and spectral information and, ultimately, detector responses that could be physically measured at various points on the process line. Such information could be used to optimize detector locations in support of real-time on-site monitoring to determine the enrichment levels within a process stream. The results of parametric analyses to establish expected variations for several different process streams and configurations are presented. The benefits and issues associated with both passive and active interrogation measurement techniques are also being explored.

INTRODUCTION

The U.S. Department of Energy (DOE) has provided funding to LLNL for developing tools and methods for potential U.S. use in designing and evaluating safeguards systems used in enrichment facilities [1]. The International Atomic Energy Agency (IAEA) is also assessing the needs and capabilities necessary to and efficiently safeguard enrichment plants. An IAEA hosted technical meeting in Vienna on April 18-22, 2005 aimed at further strengthening inspection and verification approaches applied to uranium enrichment activities.

The current efforts focus on providing continued information that can be used in identifying the placement and effectiveness of safeguards in protecting against the possible diversion of attractive materials and unauthorized activities at enrichment plants. It is part of a multi-

laboratory DOE project, following on from an earlier examination of possible safeguards for natural uranium conversion plants [2, 3].

Based on earlier work, a tool suite has been put together for safeguards analysis, the Lawrence Livermore National Laboratory LLNL (LLNL) Integrated Safeguards System Analysis Tool (LISSAT) [4]. LISSAT provides a framework for performing systems analysis and evaluating the effectiveness of a safeguard system for a nuclear fuel cycle facility. As a part of the LISSAT suite of tools, a development effort has been undertaken to build a set of routines that can be utilized anywhere in the simulation to identify the neutron and gamma flux and associated detector responses. The intent of this effort is to aid in the placement of radiation detection instrumentation to most effectively identify when diversion of material is taking place.

In a previous paper [5], we described efforts undertaken to develop a routine to provide neutron levels around process piping and feed and product containers. In the present paper, current efforts and results are described that cover the development of a routine that provides gamma flux levels around process lines anywhere in an enrichment facility.

DERIVATION OF RADIATION SOURCE TERM

The approach of the current efforts is to provide a simple set of algorithms that can provide reasonable values for both the neutron and gamma flux outside a line or container carrying either UF_6 gas or UF_6 as a solid or liquid. These algorithms are subsequently incorporated into the LISSAT suite of tools to model a uranium enrichment plant. The neutron routine previously presented [5] indicated that variations in enrichment levels would be detectable due to neutrons emanating from assumed product containers. However neutron levels emanating from process lines did not lend themselves to easy detection of changes in enrichment levels with process lines. This was due to the low gas density and small pipe diameters employed at enrichment facilities resulting in a low (α,n) production in the gas before the alphas were lost to the pipe walls, and because of interference from neutrons generated at other locations. Consequently efforts were devoted to develop an initial gamma routine to provide gamma flux levels around process piping that would indicate the degree of fluctuations with respect to changes in the enrichment levels. The intent of the development efforts is to allow one to predict the radiation fields that might be expected under certain diversion scenarios to support the identification and placement of appropriate radiation detectors in the field. At the present time, efforts are aimed at the determination of the radiation fields that might be seen by a detector, follow-on efforts would correlate the expected radiation fields with the anticipated detector responses.

The determination of the radiation fields was broken up into two basic parts. The first part was to quantify the neutron source term and resulting flux depending upon the physical geometry encountered. The results of these initial efforts were previously reported [5]. The second part of the effort as discussed in this paper is to quantify the gamma source term and resulting flux again as a function of the basic geometry encountered. In both instances, as the application is for a uranium enrichment plant wherein numerous pipes exist to transport material within the process and both feed and product containers were cylindrical, the source was assumed to be represented by a cylinder of varying diameters and lengths. Since the enrichment levels vary, depending upon where in the process one might be performing an interrogation, the source terms for both the neutrons and gammas were defined for each uranium isotope separately. Once defined, the

source term for any mix (i.e., level of enrichment) could be defined by varying the source strengths from each uranium isotope according to its percentage in the overall mix.

Gamma source term and flux determination

Gammas arise due to the decay of the various uranium isotopes, the subsequent decay of the daughter products and that due to spontaneous and neutron induced fission and the subsequent decay of the fission products. The gamma source term was based on the LLNL computer code GAMGEN [6] and NUREG/CR-5550 [7]. The source term included both the specific activity and energy spectrum from each radionuclide ^{234}U, ^{235}U, and ^{238}U. GAMGEN accounts for decay of both parent and subsequent daughters but does not account for either spontaneous or induced fission gammas or those of the fission products produced. However since the half-life for spontaneous fission is much less than that of decay, both spontaneous and neutron induced fission gammas are neglected. It was assumed that daughter products would not be volatilized when the feed container was heated and hence the amount of time from process filling of the feed container to arrival at the enrichment plant could be neglected. The amount of time the uranium hexafluoride gas would remain within the enrichment process piping was taken as an hour and hence an hour of decay time was used to determine the gamma source strength using GAMGEN. The source strength was given as specific activity, gammas emitted per unit time and mass (γ/cm^2-s). While most of the known gamma lines were tracked, those given in Table 1 were those tabulated.

Table 1 Gamma Lines Selected for Tabulation

Radionuclide	Associated Gamma Energies (MeV)			
^{234}U	0.45497	0.50353		
^{235}U	0.09336	0.14376	0.18572	0.20531
^{238}U	0.09235	0.09278	0.11280	0.76641

The gamma flux at a point perpendicular to a distance D from the midpoint of a pipe of length L containing a gamma source S_i per unit length of pipe has been treated as that of a line source as given by Equation (1). This was felt to be a good approximation for the types of gas densities and pipe sizes of interest. The adjustment factor ADJ_i is a correction factor that accounts in part for the finite source behavior of the pipe. The adjustment factor was determined by comparing the ideal line source behavior with that determined from various MNCP [8] analyses. The source term, S_i, per isotope i, as given by Equation (2), is calculated based on the pipe diameter d, the density of UF_6 gas within the pipe ρ, the fractional isotopic mix f_i , and the specific gamma activity SA_i. The determination of the total gamma flux is based on a weighted average of the flux contributions from each of the uranium nuclides as given by Equation (3). The approach utilized is accurate within a few percent up to UF_6 densities on the order of 0.01 g/cm^3. Densities greater than 0.01 g/cm^3 begin to vary from a simple line source due to self-absorption as shown in Figure 1. As can be seen by examining Figure 1, the flux starts to vary as the gas density approaches 0.01 g/cm^3 indicating the point at which self absorption begins to become important.

$$\varphi_i = \frac{S_i \cdot \arctan\left(\frac{L}{2D}\right)}{2\pi D} \cdot ADJ_i \qquad (1)$$

$$S_i = \frac{f_i \cdot \rho \cdot \pi \cdot d^2}{4} \cdot SA_i \tag{2}$$

$$\phi = \sum_{i=1}^{3} \varphi_i \tag{3}$$

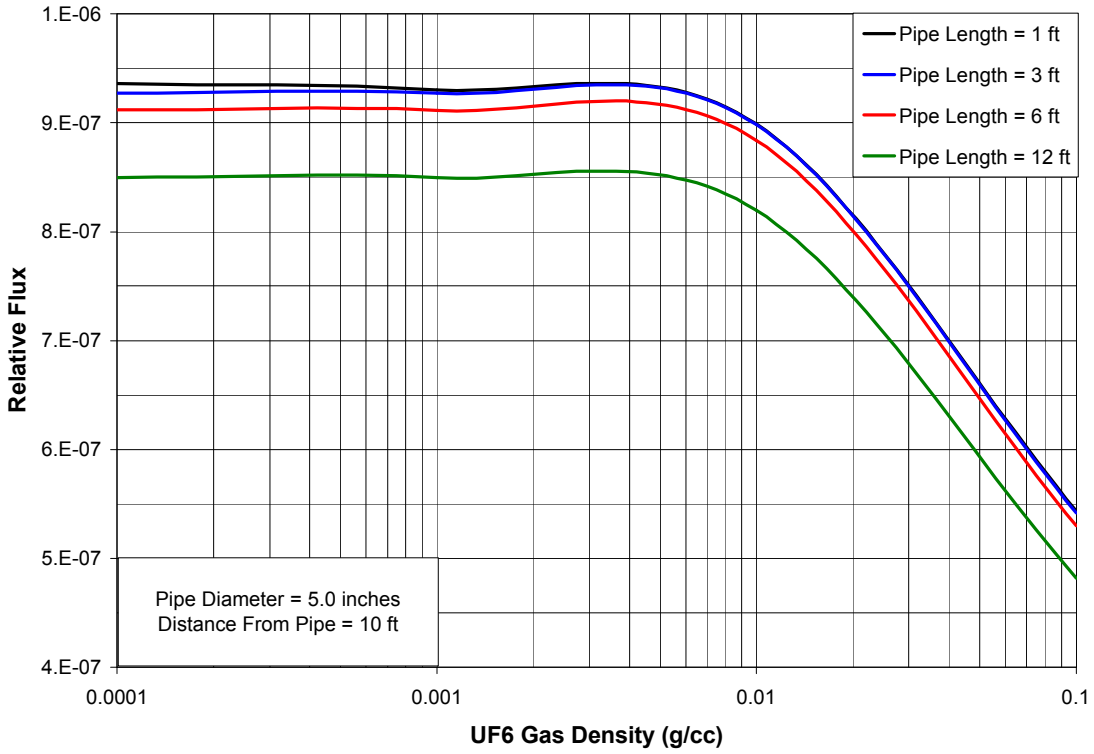

Figure 1. Effect of gas density on relative gamma flux.

The relative Gamma Flux (cm^{-2}) in Figure 1 is the ratio of the flux at a defined point in units of (γ/cm^2-s) divided by the total gamma emission rate contained in the source in units of (γ/s). The relative flux decreases with pipe length because the ends of the pipe are further from the defined point and decreases with increasing gas density due to the increase of self absorption within the gas.

Results of Gamma Source Term Modeling

Applying the expressions given in Equations (1), (2) and (3), Figure 2 shows the gamma flux for various levels of enrichment as a function of distance from the center of a 1 foot long section of pipe having a diameter of 0.5 inches and a UF_6 density of 0.001 g/cm^3. As the level of enrichment increases, the source term increases, as the specific activity of ^{238}U is the lowest of the 3 naturally occurring uranium isotopes. Hence, the total gamma flux increases.

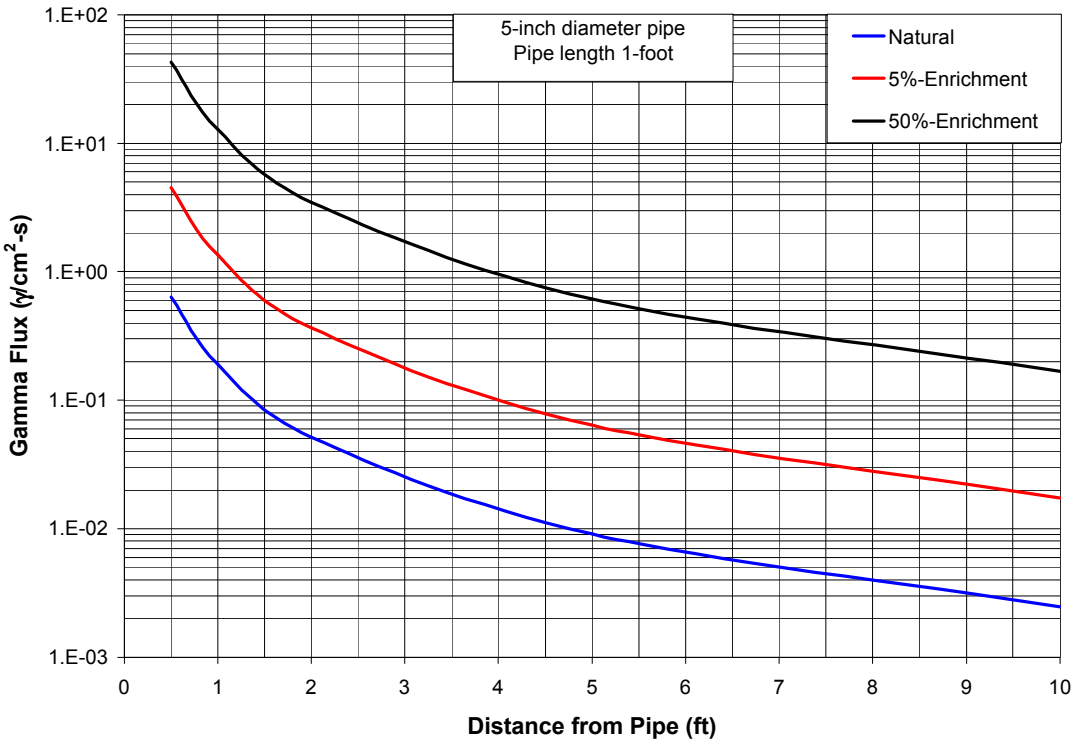

Figure 2. Gamma flux versus ^{235}U enrichment at 1 ft from a 1-ft long ½-inch pipe

In incorporating the routine into the LISSAT suite of tools, several additional algorithms were written so as to extract the necessary information from the enrichment plant model required to use the radiation source term routine. In particular, the gas density is not a property that the modeling code calculates. However, the pressure at which the system is operating is in principle known. As a consequence, Equation (4) was written, utilizing the ideal gas law, to compute the density ρ in g/cm^3 of UF_6 gas given the line pressure P in bars and ambient or operating temperature T in centigrade.

$$\rho = \frac{4.234 \cdot P}{(T + 273.16)} \tag{4}$$

For example, at T = 25 °C and P = 0.050 bars, ρ = 0.00071 g/cm^3.

The following section illustrates the results obtainable from the modeling code with the radiation source term routine as incorporated in LISSAT.

SIMULATION RESULTS

The gamma flux module was integrated into the Extend simulation model for the generic enrichment facility [4]. The measurement point for the gamma flux is assumed to be about 1 ft from a process line containing the diverted UF_6 having an enrichment of 50% ^{235}U. Figure 3 shows the increase in gamma flux emanating from the process line as the section of pipe

collimated increases and hence a detector's view. The detector would be located as close to the process line as reasonably possible so as to maximize the gamma flux. At the same time, the view would be collimated to minimize the contributions due to scattering and that due to other process lines. As can be seen by examining Figure 3, a reasonable flux can be obtained by viewing roughly 2 feet of pipe.

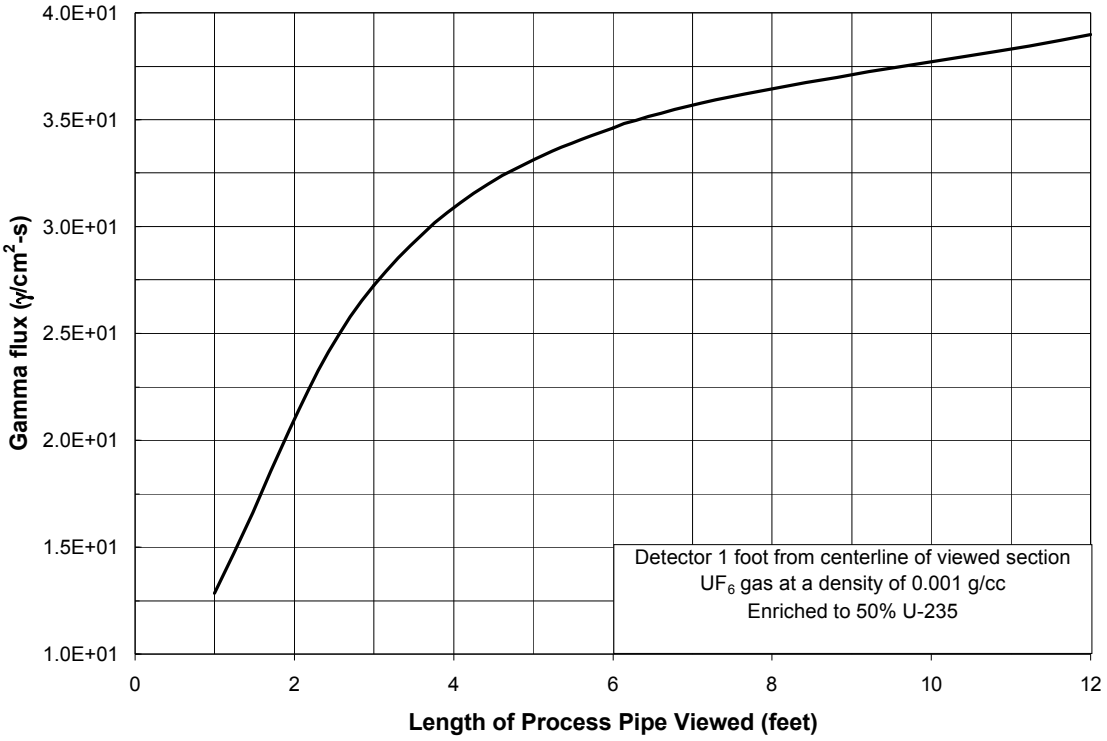

Figure 3. Gamma flux at 1 ft from a process line with UF_6 gas at 50% enrichment.

FUTURE EFFORTS

The initial approach of our efforts was to develop a fast running routine that would provide both neutron and gamma fluxes. At the present time, efforts are continuing to increase the range of the gamma routine to provide gamma flux levels at solid densities so that the flux from both feed and product containers can be determined. In addition, the routine is to be modified to provide flux levels for the specific lines previously given in Table 1. Plans are to couple the flux and energy spectrum into both a neutron and gamma detector to provide its response.

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