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# **Modeling Efforts to Aid in the Determination of Process Enrichment Levels for Identifying Potential Material Diversion**

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## **ABSTRACT**

Efforts have been under way at Lawrence Livermore National Laboratory (LLNL) to develop detailed analytical models that simulate enrichment and conversion facilities for the purpose of aiding in the identification of possible areas where material diversion could occur as part of an overall safeguards strategy. Operation of an enrichment process for manufacturing commercial reactor fuel presents proliferation concerns including both diversion and the potential for undeclared enrichment to make weapons grade material. Inspections by the International Atomic Energy Agency (IAEA) are designed to provide assurance that such diversion is not occurring through, among other things, visual examination of the facility and taking specific measurements such as the radiation fields outside of various process lines. Our current effort is aimed at developing algorithms that would be incorporated into the current process models that would provide estimates of both neutron and gamma radiation fields outside any process line for the purpose of determining the most effective locations for placing in-plant monitoring equipment. These algorithms, while providing dose and spectral information, could also be designed to provide 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. Based upon these results, the capability of a sodium iodide (NaI(Tl)), high-purity germanium (HPGe), or neutron detection system is being investigated from the standpoint of their viability in quantitatively measuring and discerning the enrichment of in-process material. The benefits and issues associated with both passive and active interrogation measurement techniques are also discussed.

## **INTRODUCTION**

The U.S. Department of Energy (DOE) is interested in 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 is also continuing to review needs, capabilities, and efficiency in safeguarding enrichment plants. The IAEA hosted a technical meeting in Vienna on April 18-22, 2005 with the aim of further strengthening its inspection and verification approaches applied to uranium enrichment activities.

The present applied research focuses on providing additional information that can be used in identifying the placement and effectiveness of safeguards in protecting against the possible diversion of attractive material and unauthorized modes of use 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 the earlier work, we have put together a tool suite for safeguards analysis, the Lawrence Livermore National Laboratory LLNL Integrated Safeguards System Analysis Tool (LISSAT) [4]. It is a framework for performing systems analysis for 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 model 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 this paper we describe the status of our current development efforts to define the neutron and gamma flux emanating from a process line anywhere in an enrichment facility.

## **DERIVATION OF RADIATION SOURCE TERM**

The approach being utilized 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<sub>6</sub> gas or UF<sub>6</sub> as a solid or liquid. These algorithms are being incorporated into the LISSAT suite of tools to model a uranium enrichment plant. The intent is to look at the radiation fields that might be expected under certain diversion scenarios to support the identification and placement of appropriate radiation detectors in the field. While this first stage deals with the determination of the radiation fields that might be seen by a detector, follow-on efforts will correlate the expected radiation fields with the anticipated detector responses.

The overall problem of determining 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 second part was to quantify the gamma source term and resulting flux again as a function of the basic geometry encountered. In both instances since the application was 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, lengths, and thicknesses along with two material types, aluminum and stainless steel. 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.

### ***Neutron source term and flux determination***

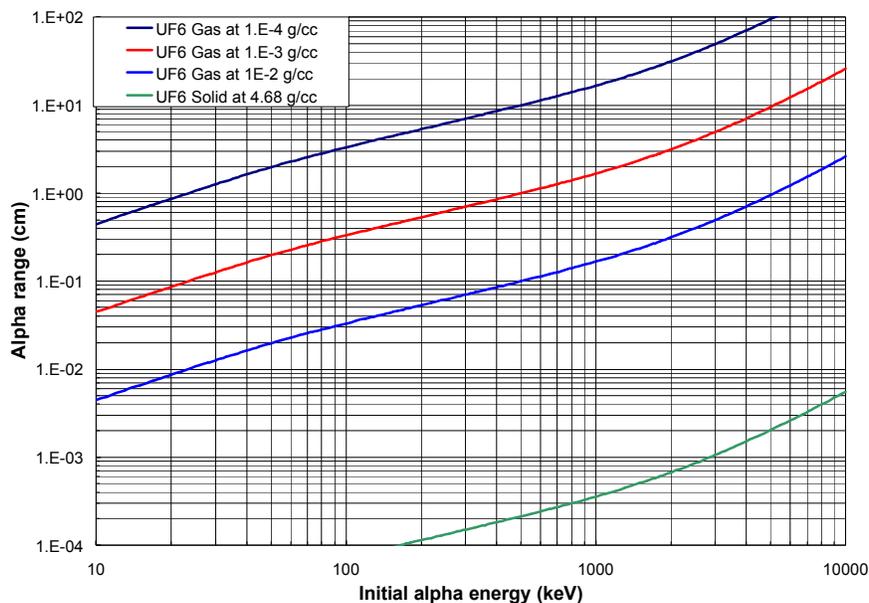
Neutrons can arise due to several sources and interactions. In uranium, neutrons can be generated due to spontaneous fission and neutron induced fission. In the latter case, the overall neutron source might be increased if the degree of multiplication, based on enrichment level and geometry, is significant. As uranium decays by alpha emission, another source of neutrons is due to the potential for alpha-n reactions. Such alpha-n reactions take place in the lighter low-Z materials. Table 1 provides a listing of low-Z elements that give rise to this phenomenon. Of particular interest, in the case of enrichment, is the fact that the uranium is handled as a gas in the form of UF<sub>6</sub>, uranium hexafluoride. Fluorine in turn is a light element that reacts with an alpha to give a neutron. Because the spontaneous fission rate is so low in all the uranium isotopes of

interest,  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , the alpha-n reaction is one of the largest contributors to the neutron source term. Further, neutrons can also be generated as a result of the alpha-n reaction occurring in the pipe or container material through which the  $\text{UF}_6$  gas is confined or solid stored.

**Table 1 Some Radionuclides that Generate Neutrons as a Result of an Alpha-n Reaction**

$^7\text{Li}$	$^{13}\text{C}$	$^{19}\text{F}$	$^{25}\text{Mg}$	$^{30}\text{Si}$
$^9\text{Be}$	$^{14}\text{N}$	$^{21}\text{Ne}$	$^{26}\text{Mg}$	$^{31}\text{P}$
$^{10}\text{B}$	$^{17}\text{O}$	$^{22}\text{Ne}$	$^{27}\text{Al}$	$^{37}\text{Cl}$
$^{11}\text{B}$	$^{18}\text{O}$	$^{23}\text{Na}$	$^{29}\text{Si}$	

In the case of a gas, the alpha range can be quite large especially at the reduced pressures one might encounter in a gaseous diffusion or centrifuge plant. The range of an alpha as a function of gas density is given in Figure 1 based on results from the TRIM code [5]. The rate at which neutrons were generated as a result of the alpha-n reactions was based on the code SOURCES-4C [6]. The code SOURCES-4C computes the reaction rates on the assumption that the medium is infinite with respect to the alpha range. While this is a good assumption for  $\text{UF}_6$  in the solid or liquid phase, as the alpha particle has a range on the order of tens of microns, it is not a good assumption in the gaseous phase where, due to the working pressures of an enrichment plant, the gas density can be on the order of  $0.1 \text{ mg/cm}^3$  and the range of an alpha particle can be on the order of 1 m. The alpha energies resulting from the uranium isotopes examined are given in Table 2 [7]. As the alpha traverses through the  $\text{UF}_6$  medium, it steadily loses energy. The probability that the alpha will undergo an  $(\alpha, n)$  reaction is nonzero down to the threshold energy, corresponding to a threshold path length. An initial adjustment has been made to account for the fact that the  $\text{UF}_6$  media is non-infinite by dividing the radius of the process pipe by the sum of the radius of the process pipe and the threshold path length of the alpha. The approach is illustrated in Figure 2. A more detailed analysis involving the modeling of the alpha tracks,

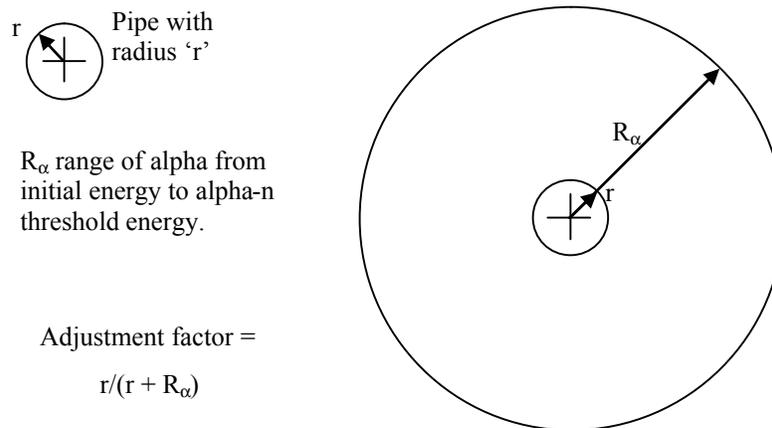


**Figure 1 Alpha Range in  $\text{UF}_6$  as a Function of Density**

trajectories, interactions in such a finite media, and decreasing cross section with decreasing energy [6] to determine the actual interaction rate is planned.

**Table 2 Relative Intensity and Energy of Prominent Uranium Alpha Particles**

Radionuclides					
<sup>234</sup> U		<sup>235</sup> U		<sup>238</sup> U	
Peak Alpha Intensity and Energy					
(%)	(MeV)	(%)	(MeV)	(%)	(MeV)
72	4.773	4.6	4.597	77	4.195
28	4.722	3.7	4.556	23	4.147
		4	4.415		
		57	4.396		
		18	4.366		
		5.7	4.216		
Average	4.77	Average	4.41	Average	4.19



**Figure 2 Adjustment Factor to Account for Finite Dimensions of Piping**

The neutron flux at a point that is a perpendicular distance  $D$  from the midpoint of a pipe having a length  $L$  due to a neutron source  $S$  contained within a pipe has been treated as that of a line source and is given by Equation (1). This was felt to be a good approximation for the types of gas densities and pipe sizes of interest. The source term,  $S$ , is calculated based on the pipe diameter  $d$ , the density of gas within the pipe  $\rho$ , the fractional isotopic mix  $f_i$ , the alpha-n neutron source  $AN_i$  and spontaneous fission neutron source  $SF_i$  (from SOURCES-4C), and an adjustment factor to account for the finite nature of the source  $ADJ_i$  as given in Equation (2). The summation in Equation (2) is over all three uranium isotopes and the determination of the adjustment factor is as given in Figure 2. The approach utilized is somewhat less accurate when it comes to the solid and liquid phases of  $UF_6$  because of the possible increase in neutron multiplication and absorption. Again, a more detailed analysis is being carried out using the code MCNP [8] to account for both the multiplicative and absorption effects. The results from these analyses will be used to refine the current algorithms.

$$\varphi = \frac{S \cdot \arctan\left(\frac{L}{2D}\right)}{2\pi D} \quad (1)$$

$$S = \frac{\rho \cdot \pi \cdot d^2}{4} \sum_{i=1}^3 f_i \cdot ((AN_i \cdot ADJ_i) + SF_i) \quad (2)$$

### ***Gamma source term and flux determination***

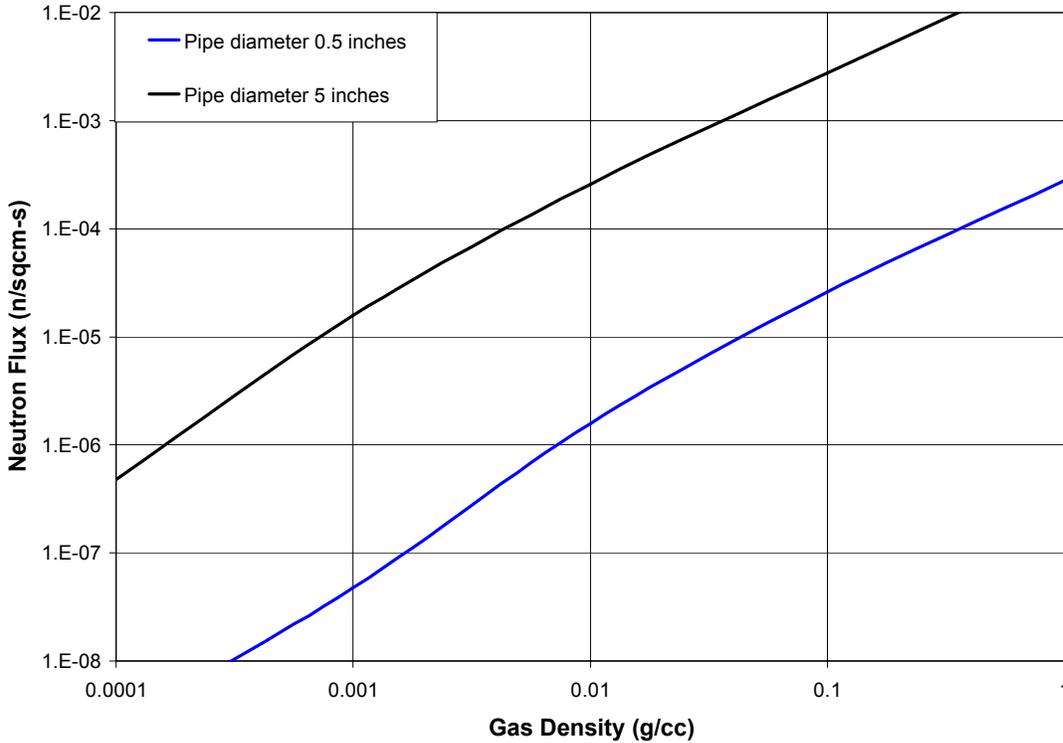
In the case of gammas, the energy spectrum from each radionuclide  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  was determined using the LLNL code GAMGEN [9]. The current analysis has focused on virgin material, defined as material with one hour of decay. Unlike the neutrons, the lower energy gammas can be significantly attenuated by the wall thickness of the pipe. Depending on the density of  $\text{UF}_6$  in the pipe or container, the degree of self attenuation by the source might be neglected. For example, at a  $\text{UF}_6$  density of  $0.001 \text{ g/cm}^3$ , the degree of self-attenuation of a 100 keV gamma in a 5" pipe can be calculated to be about 9%. Thus at typical gaseous densities the self shielding effect can reasonably be neglected. However, the attenuating effect of the pipe wall needs to be taken into account. At liquid and solid densities in the range from 3 to  $5 \text{ g/cm}^3$ , roughly 99% of the 100 keV gammas would be attenuated after passing through roughly 50 mils of the source material. To properly account for the self attenuation and that due to the walls of the pipe or container, the gamma flux is being determined through the use of the code MCNP. A range of pipe diameters, densities, and lengths are being analyzed to determine the gamma flux at various distances. While the input covers the entire spectrum of gamma energies, only selected gamma peaks have been chosen to be utilized in the routines being developed. The current energies of the gamma peaks selected are summarized in Table 3. Results of the gamma analyses will be presented at a later time.

**Table 3 Gamma Lines Selected for Tracking**

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

### ***Results of Neutron Source Term Modeling***

Applying the expressions given in Equations (1) and (2) and Figure 2 for a gas of varying density is shown in Figure 4. As the density of the gas increases, the source term increases first because of the increase in activity but also because the alpha range is decreasing and hence the adjustment factor to account for the finite nature of the geometry with respect to the range of the alpha particles begins to approach unity. As currently written, the routine uses a density of  $4.68 \text{ g/cm}^3$  for solid  $\text{UF}_6$  which will be modified in the future to allow variations there as well.



**Figure 4 Neutron Flux versus UF6 Gas Density at 3 ft from a 12 ft 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 known. As a consequence, Equation (3) was written, utilizing the ideal gas law, to compute the density  $\rho$  in  $\text{g/cm}^3$  of  $\text{UF}_6$  gas given the line pressure  $P$  in bars and ambient or operating temperature  $T$  in centigrade. In the case where a container is being filled and the material solidified therein, an effective diameter is calculated based on the expression given by Equation (4) where the effective diameter  $d_{eff}$  is in inches, the mass flow rate  $\dot{M}$  is in units of  $\text{KgU/h}$  and  $L$  the length of the container is in feet.

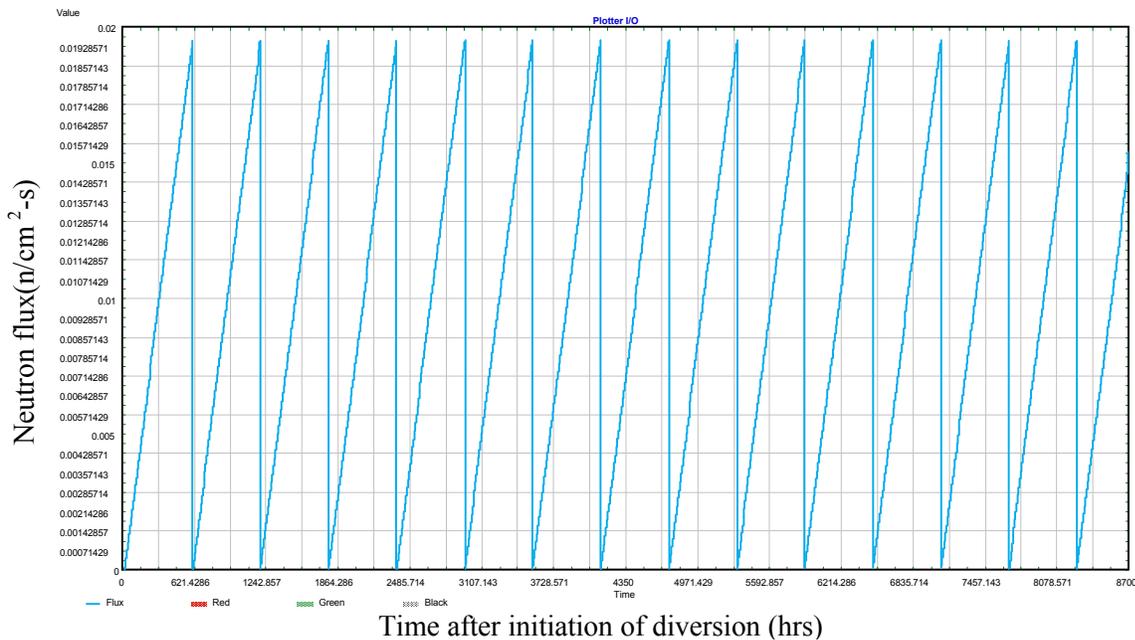
$$\rho = \frac{4.234 \cdot P}{(T + 273.16)} \quad (3)$$

$$d_{eff} = \sqrt{\frac{\dot{M} \cdot t}{L}} \quad (4)$$

The following section illustrates the results from the modeling code with the radiation source term routine incorporated and exercised.

## SIMULATION RESULTS

The neutron flux module was integrated into the Extend simulation model for the generic enrichment facility [4]. The measurement point for the neutron flux is assumed to be about 10 ft from the cylinders containing the diverted LEU. About 2000 Kg of LEU were assumed to be diverted by skimming from one cascade during the year. The cylinder was assumed to be a 12A capable of holding about 141 Kg of LEU. It takes about 600 hrs. to fill one container with an associated container length of about 3 feet. Figure 5 shows how the neutron flux emanating from the container varies over time as the container is being filled. The saw tooth nature of the curve is a result of the continuous change out of containers as each one becomes filled over the diversion time of a year. When a cylinder is full, the peak neutron flux is about 0.02 neutron/cm<sup>2</sup>-sec.



**Fig. 5. The neutron flux at 10 ft from a 12A container as reported by the neutron flux module over a year.**

## FUTURE EFFORTS

The initial approach of our efforts was to develop a fast running routine that would provide both neutron and gamma fluxes and spectra. While development continues on the gamma portion of the routine, the initial portion of the neutron flux calculation has been completed. Continuing efforts include refining the neutron calculation to properly handle attenuation and multiplication in high density materials, solids and liquids, and high enrichment scenarios. In addition, the coupling of the flux and energy spectrum to both a neutron and gamma detector so as to model the detector response is planned. The detector response of a He-3 neutron detector would be modeled using an MCNP model of the detector. The detector response for gammas would most likely use the code GADRAS for a high purity germanium detector [10, 11].

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