MEASUREMENT METHODOLOGY OF THE FISSILE MASS FLOW MONITOR FOR THE HEU TRANSPARENCY IMPLEMENTATION INSTRUMENTATION IN RUSSIA

Taner Uckan, Rick Oberer, and Josè March-Leuba
Oak Ridge National Laboratory,[*] *P. O. Box 2008, Oak Ridge, TN 37831 USA*

Danny Powell, and Jim Sumner
BWXT Y-12, L.L.C., *P. O. Box 2009, Oak Ridge, TN 37831 USA*

Edward Mastal
U.S. Department of Energy, *19901 Germantown Rd, Germantown, MD 20874 USA*

Abstract

The highly enriched uranium (HEU) Transparency Agreement between the U.S. and Russian Federation (RF) requires implementation of transparency measures in the Russian facilities that are supplying product low enriched uranium (LEU) to the U.S. from down blended weapon-grade HEU material. To satisfy the agreement's non-proliferation objectives, the U.S. DOE is implementing the fissile mass flow monitor (FMFM) instrumentation developed by Oak Ridge National Laboratory. The FMFM provides unattended non-intrusive measurements of ^{235}U mass flow of the uranium hexafluoride (UF_6) gas in the process lines of HEU, the LEU blend stock, and the resulting lower assay product LEU (P-LEU) that is used for U.S. reactors. The instrumentation continuously traces the HEU flow through the blending point to the product LEU, enabling the U.S. to verify HEU material down blending. The FMFM relies on producing delayed gamma rays emitted from fission fragments carried by the UF_6 flow. A thermalized californium-252 (^{252}Cf)-neutron source placed in an annular sleeve filled with moderator material that surrounds the pipe is modulated by a neutron absorbent shutter to induce fission in UF_6. For this technique to be effectively applicable the average range of resulting fission fragments in the UF_6 gas must be smaller than the pipe diameter. The fission fragment range can be very large in low-density materials. Therefore, a methodology has been developed to determine the fission fragment range and its distribution to assess the fraction of the fission fragments that will remain in the flow; this methodology is the primary topic of discussions in this paper.

Introduction

The government-to-government HEU Purchase Agreement signed in February 1993 between the U.S. and the RF provides transparency measures be implemented at U.S. and Russian nuclear facilities processing uranium subject to the agreement. Moreover this agreement provides for the monitoring of the down blending of HEU at an assay of ~90% with blend stock LEU at an assay of ~1.5% to produce reactor-grade material at an assay of ~4%, P-LEU, to be used in U.S. nuclear power plants. The Ministry of the RF for Atomic Energy (MINATOM) and the U.S. Department of Energy have agreed on implementing transparency measures at the Ural Electrochemical Integrated Plant (UEIP) at Novouralsk, Russia, and the Electrochemical Plant (ECP) at Zelenogorsk, Russia. These transparency measures include the installation of the Blend Down Monitoring System (BDMS) to monitor the enrichment and fissile mass flow of the HEU blending processes at UEIP and at ECP. The BDMS has been developed to provide unattended and continuous monitoring of the HEU blending operations at these Russian facilities. The BDMS consists of the Enrichment Monitor (EM) developed by the Los Alamos National Laboratory and the FMFM, and in Figure 1 the BDMS locations are shown on the legs of the HEU blending tee.

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[*] Managed by UT-Battelle for the U.S. Department of Energy under contract DE-AC05-00OR22725.

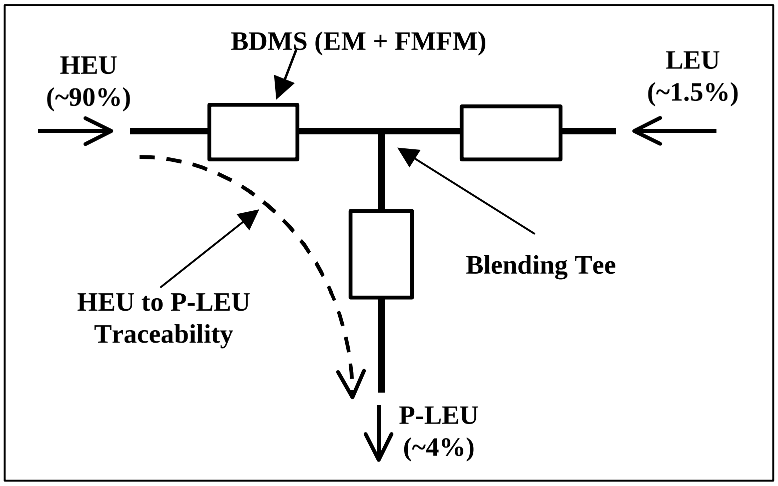

Figure 1. The Blend Down Monitoring System (BDMS) installed on a HEU blending tee.

In addition, the FMFM also traces fission products generated in the HEU stream through the blending operation into the P-LEU stream, thus confirming the HEU material down blending process as schematically indicated in Figure 1. This HEU traceability gives U.S. monitors significant confidence that the HEU is indeed being blended into a lower assay material. The first BDMS was successfully implemented and is operational at the UEIP since February 1999.

Operational Principles of Fissile Mass Flow Monitor

The FMFM measures the fissile mass flow of the UF_6 gas in the HEU, the P-LEU, and the LEU blend stock process lines of the blending tee (see Figure 1) non-intrusively. To do this, ^{235}U fissions are induced in the UF_6 fissile flow stream by a thermalized and modulated ^{252}Cf-neutron source placed on each process line as shown in Figure 2.

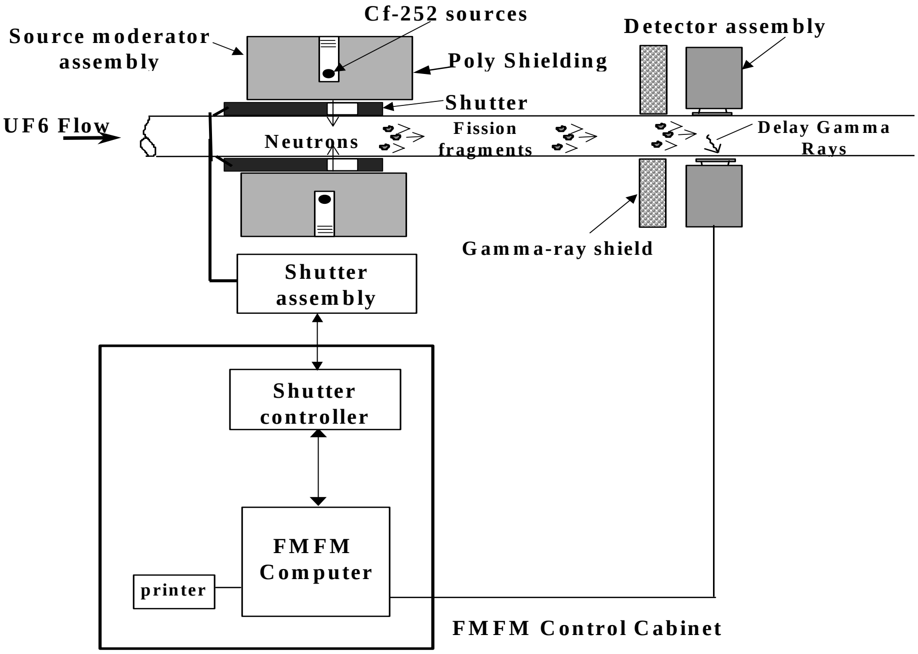

Figure 2. Operational principle and the major components of the Fissile Mass Flow Monitor.

The induced fissions are time modulated using a neutron-absorbing shutter to create a time signature in the UF_6 gas. A set of gamma ray detectors, located downstream of the source, measures delayed gamma rays emitted by the resulting fission fragments. Then, the FMFM determines the fissile mass flow rate from two independent measurements: (1) the observed delay in the time correlated measurement between the source modulator and the detector signal provides the velocity of UF_6, and (2) its amplitude is related to the ^{235}U concentration in UF_6. The HEU material traceability is accomplished by detecting the presence of the time-modulated fission fragment signal in the P-LEU leg created in the HEU leg of the blending tee as shown in Figure 1. An on-line computer controls the source modulator, processes acquired detector data, and reports results.

To predict the detector response from the measurements of the gamma rays resulting from the fission fragment production downstream of the source, it is necessary to estimate (a) the fraction of fission fragments (ε_f) that remain in the UF_6 gas following an induced fission by the Cf-neutron source, and this is the main topic of this paper, (b) the transport of the fission products in the pipe, and (c) the rate of decay of the fission products produced in the UF_6 gas. The details of the FMFM models employed for the last two topics were discussed in earlier publication [1].

Problem and Objective

The concentration of ^{235}U fissile material in UF_6 flowing in a process pipe is estimated by measuring gamma rays emitted from fission fragments carried along by the flowing gas. The fission fragments in a low density ($\sim 5 \times 10^{-4}$ g/cm^3) UF_6 have a considerable range at a typical process pressure of ~40 Torr (see Table 1). Because of this possible long range, some of the fragments are lost in the pipe wall. These fragments are therefore not available to emit gamma rays down stream. Given fixed pipe dimensions and the UF_6 gas pressure, the probability that a fission fragment will be lost depends on the probability density function (PDF) of fragment ranges. This PDF was previously estimated from *Nuclear Data Tables* (NTD) [2,3]. The disadvantage of this approach is that data is available for only a few of the many fission fragments, data is not available for the actual stopping media of UF_6 gas, average values rather than PDFs are given and the data must be adjusted for straggling effects. The primary goal of this paper is to reconstruct the PDF of the fragment range using TRIM (Transport of Ions in Matter) code (STRIM 2000.39) [4] and compare the result with the previous estimate [3].

TRIM Code and Calculations

The TRIM is the most comprehensive of the group of programs included in the SRIM (Stopping and Range of Ions in matter) code package. It calculates the transport of ions in matter using a quantum mechanical treatment of ion-atom collisions. SRIM resulted from the original work by J.P. Biersack on range algorithm [5], and work by J. F. Ziegler on stopping theory [4]. The energetic charged particle, a fission fragment in this application, is referred to as an “ion” in the TRIM code. The material, through which the ion is transported, which is UF_6 gas in this case, is referred to as the “target” in the code.

Range Distribution

TRIM tallies a histogram of the depth in the target, which the ions stop. This is the final position of the fragment in the direction of the initial velocity. Examples of these histograms are shown in Figure 3 for the fission fragment ^{90}Kr. Although radial range might be more appropriate, it is found that the default TRIM histogram to be a reasonable estimate for the range PDF for a fission fragment. For example, in the case of the fission fragment ^{80}Ge, the average depth was 10-cm in UF_6 at pressure of 40 Torr. The average radial range was 10.02-cm. The convenience of the depth rather than the radial range outweighed the less than quarter percent error in the average. It might also be suggested that the maximum depth or range is more appropriate than the final depth. Again the two are virtually indistinguishable. An ion nearly always stops very near the maximum depth. Recoils are very rare events. Furthermore, recoil is at least as likely to return a fission fragment to the gas from the pipe wall as not.

A. Range as Function of UF_6 Pressure

Full TRIM calculations were performed for ^{90}Kr in UF_6 gas at a pressure range of 20-50 Torr. This was done to determine if a single pressure was sufficient to represent the range PDF. The ion energy of 99.8 MeV was used for this comparison representing an avareage energy of light fission fragments [6]. The range of pressures and corresponding UF_6 densities [7] are shown in Table 1.

Table 1. **UF_6 Density at Various Pressures at 25°C**

Pressure (Torr)	Density (10^{-4}g/cm^3)
20	3.8
30	5.7
40	7.6
50	9.5

The range PDFs for the different pressures are shown in Figure 3. It was found that the PDF scaled according to the inverse of UF_6 gas pressure. This scaling relation is given in Figure 4, which is obtained from the most probable range for the given pressure.

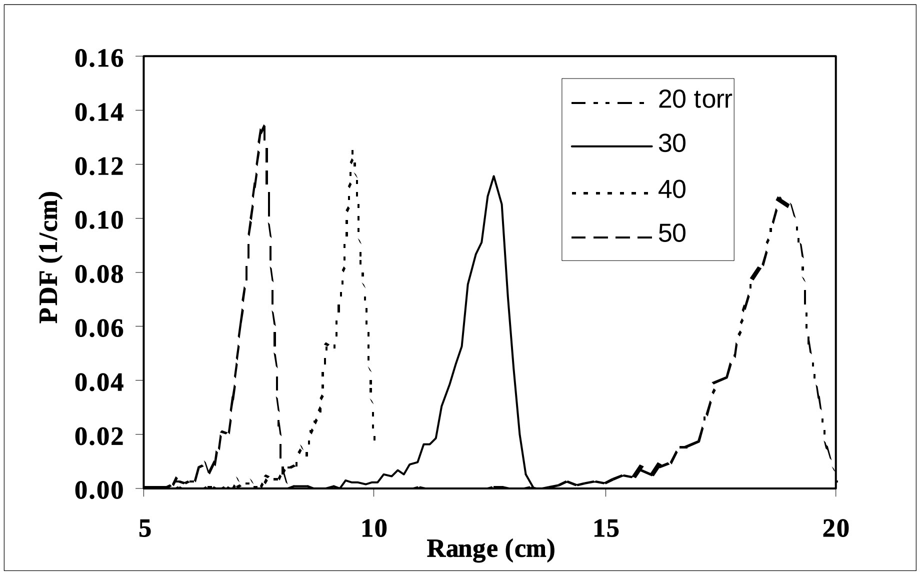

Figure 3. Range PDFs for ^{90}Kr in UF_6 at various UF_6 gas pressures.

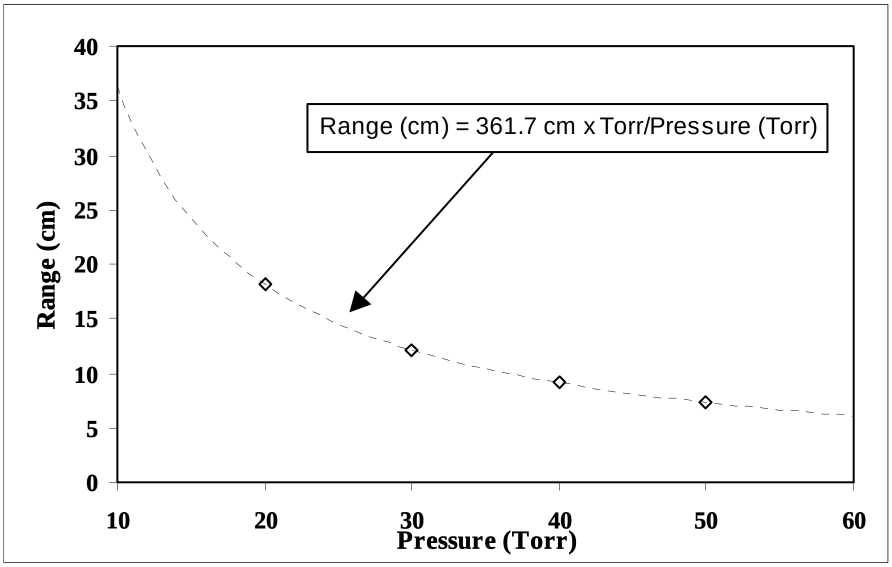

Figure 4. Average ranges for 99.8 MeV ^{90}Kr in UF$_6$ as a function of pressures.

B. Selection of Fission Fragments

There are over 700 fission fragments from the fission of ^{235}U [8], and it is impractical to calculate the range distribution for all of them. Instead fourteen representative fission fragments were chosen. These fourteen are listed in Table 2 along with their relative yield and energy. The range of a fission fragment depends on its energy (E), mass (A) and atomic (Z) numbers. In addition, a particular fragment's contribution to the over all PDF of fission fragments ranges depends on its yield. The effect of these factors will be discussed. First the effect of the Z number for a given mass will be considered in conjunction with yield considerations. Then the method for determining the energy of the fragment for the TRIM calculation will be discussed.

Table 2. Representative Fission Fragments and Their Energies

Fragment (j)	Relative Yield (%) (Y_j)	Energy (MeV)
^{80}Ge	0.32	110.5
^{85}As	3.19	106.9
^{90}Kr	14.61	103.2
^{95}Sr	16.26	99.6
^{100}Zr	15.65	96.0
^{105}Mo	2.41	92.4
^{110}Tc	0.06	88.8
^{125}Sn	0.08	77.6
^{130}Sn	4.49	74.3
^{135}Te	16.27	70.7
^{140}Xe	15.46	67.0
^{145}La	9.81	63.4
^{150}Ce	1.62	59.8

C. Range as a Function of Fission Fragment Atomic Number

The range of a fission fragment of a given mass and energy depends on the *Z* number of the fragment. Calculations were performed for the six isotopes with a mass of 90 amu in UF_6 gas at pressure of 40 Torr. The energy of the isotope was treated as independent of the *Z* number. Energy of 99.8 MeV was again used for this comparison. These isotopes along with the *Z* number, half life, percent probability of production from ^{235}U fission and average range are given in Table 3. Although the range varies considerably by *Z*, when the PDFs are scaled by yield, the variation is seen to merely broaden the range PDF slightly. This effect can be seen in Figure 5.

Table 3. Six Fission Fragment Isotopes with *A* = 90

Isotope *A* = 90	*Z*	Half Life	Production Yield (%)	Range (cm)
Se	34	0.427 sec	1.27E-02	9.52
Br	35	1.9 sec	5.53E-01	9.40
Kr	36	32.3 sec	4.40E+00	9.19
Rb-m	37	4.3 min	8.46E-01	9.04
Rb		2.6 min	1.39E-01	9.04
Sr	38	29.1 Year	7.37E-02	8.85
Y	39	2.67 day	8.97E-06	8.68

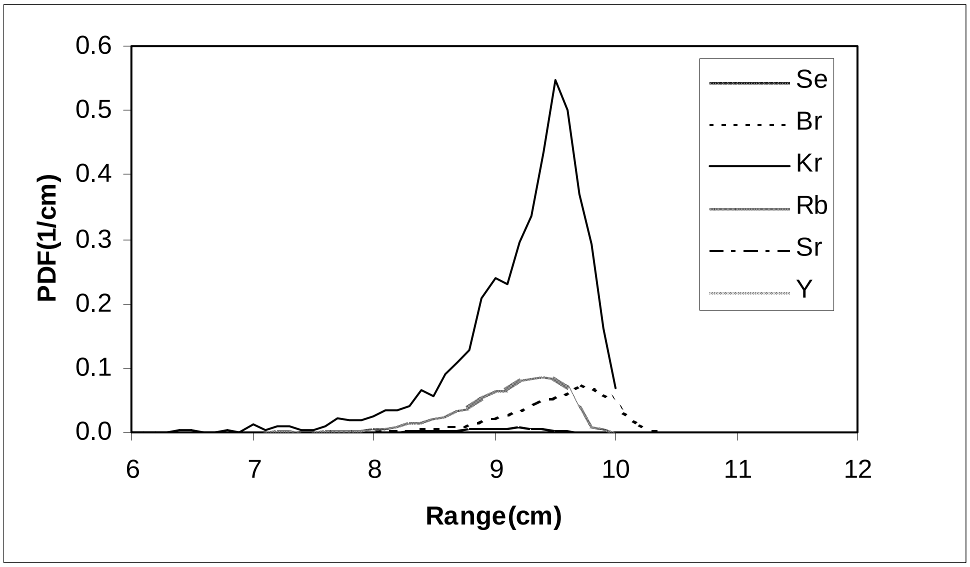

Figure 5. Range PDFs in 40 Torr of UF_6 gas for the six isotopes (*A* = 90) scaled by probability of production yield (see Table 3) from ^{235}U fission.

D. Fission Fragment Energy

Initially the energy released by fission is conserved through the kinetic energy of two fission fragments and the excitation of the two fragments. Some of this excitation energy is then released as radiation such as prompt neutrons and prompt gammas. The remainder of the excitation energy is released in subsequent gamma and beta decay. On average, 168.2 MeV goes to the kinetic energy of the fission fragments [6]. This average kinetic energy was divided between fission fragments on the basis of conservation of momentum and the resulting values are given in Table 2. This approach ignores the fact that the actual kinetic energy can vary and that the pairing of fragments as well as the number of neutrons can also vary. Range PDF$_j$ in 40 Torr of UF_6 were tallied with TRIM for each of the fourteen fission fragments (j) listed in Table 2. These PDF$_j$ was then scaled by the relative yield (Y_j) as shown in Table 2, and they are added together to form an estimate of the total range PDF = $p = \Sigma_j Y_j \times \mathrm{PDF}_j$ for ^{235}U, shown in Figure 6.

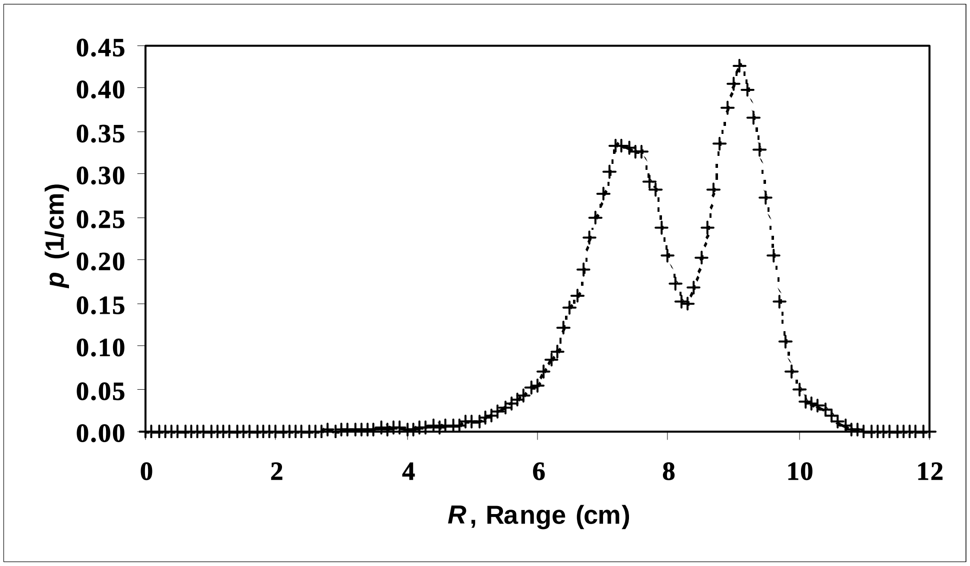

Figure 6. ^{235}U fission fragment total range PDF, *p*, obtained from the composition of range PDFs of the fragments listed in Table 2.

Monte Carlo Estimation of Fission Fragments Confined in UF_6 Flow

To determine the number of fission fragments absorbed in the pipe wall, a Monte Carlo algorithm was utilized. The steps of the algorithm are as follows: (1) Because the UF_6 density is low, the location of a fission event was selected from a uniform distribution within the pipe of diameter *D*. (2) The direction of the fission fragment was selected from an isotropic distribution. (3) The fission fragment range, *R*, was sampled from the inverse of the range cumulative probability distribution as $R = P^{-1}(\xi)$, where, *P* is the range cumulative probability distribution, $P(R) = \int_0^R p(x)dx$, and ξ is a random number uniformly distributed on (0,1). This resulting range was then compared to the distance to the pipe wall from the location of the fission event in the direction of the fission fragment. If the distance to the pipe wall exceeded the range of the fission fragment, the event was tallied (N_f) and then divided by the total number of fission fragments (N_t). This fraction, $\varepsilon_f = N_f/N_t$, of fission fragments are those confined in the UF_6 flow and eventually contribute to the delayed gamma rays at the detector located at the downstream of the Cf-neutron source. The Monte Carlo results are given in Figure 7 together with the previous estimate [3], and the analytical best fit to these data. Here the fit ε_f can be represented as $\varepsilon_f = 1/\{1 + \exp[-0.025\times(\text{pressure}\times D/51.7 - 65)]\} - 0.271$, where the UF_6 gas pressure is in Torr, and the inner diameter of the pipe *D* is in millimeters. At a typical FMFM operational pressure of 40 Torr with *D* = 100-mm pipe the fraction of fission fragments that are confined in the UF_6 flow is ε_f = 27.5% and this is about 10% lower than previous estimate [3]. The difference between the two results becomes less for higher values of pressure×*D* (see Figure 7). The fraction of fragments that remains in the UF_6 gas provides a measure of the source effectiveness of fission fragments for a yielding statistically resolvable gamma ray signal at the downstream detector for determining the ^{235}U concentration. The “source effectiveness” plays an important role for evaluating the FMFM measurement performance for a given operating UF_6 gas pressure and the process pipe size.

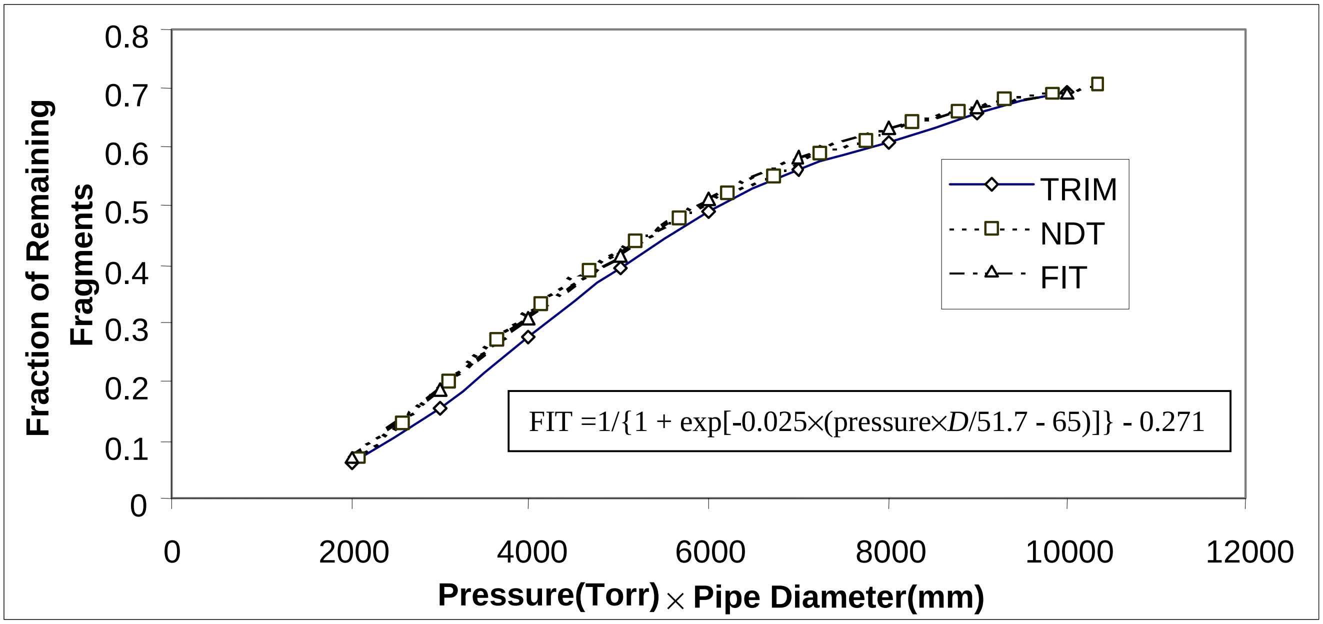

Figure 7. Fraction of fission fragments confined in UF_6 as a function of gas pressure (Torr) × inner pipe diameter (mm) estimated from TRIM and previous work using NDT [2, 3] together with the analytical best fit.

Conclusion

The fission fragment methodology presented here has been implemented in the FMFM software, which was developed to model the transport of fission fragments to interpret the measured delayed gamma ray signal from detectors located downstream of the Cf-neutron source. The TRIM code calculations have provided an independent verification of the previous estimate [3] of "source effectiveness" parameter ε_f, which was obtained from use of NDT as discussed earlier.

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