OAK RIDGE
National Laboratory

MANAGED BY UT-BATTELLE
FOR THE DEPARTMENT OF ENERGY

ORNL/TM-2003/184

^{241}AmBe Sealed Neutron Source Assessment Studies for the Fissile Mass Flow Monitor

Taner Uckan
José March-Leuba
Danny Powell
James D. White
Joseph Glaser

July 2003

UT-BATTELLE

ORNL/TM-2003/184

^{241}AmBe SEALED NEUTRON SOURCE ASSESSMENT STUDIES FOR THE FISSILE MASS FLOW MONITOR

Taner Uckan, José March-Leuba, Danny Powell, and James D. White

Nuclear Science and Technology Division

Joseph Glaser*

Presented at the Institute of Nuclear Materials Management
44th Annual Meeting, Phoenix, Ariz.
July 13–17, 2003

July 2003

Prepared by
OAK RIDGE NATIONAL LABORATORY
P. O. Box 2008
Oak Ridge, Tennessee 37831-6010
managed by
UT-Battelle, LLC
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

*U.S. Department of Energy, Germantown, MD.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

^{241}AmBe SEALED NEUTRON SOURCE ASSESSMENT STUDIES FOR THE FISSILE MASS FLOW MONITOR

ABSTRACT

In this paper the assessment studies performed on the fissile mass flow monitor (FMFM) system for possible use of ^{241}AmBe sealed neutron sources are presented. The FMFM uses ^{252}Cf neutron sources for fission activation of a UF_6 gas stream for ^{235}U fissile mass flow rate measurements. Present FMFM sources are replaced about every 2 years because the half-life of ^{252}Cf is relatively short (~2.65 years). This is a costly process, not only for the source fabrication, but also because of the associated costs of transportation, installation, and storage. In addition, the sources are calibrated with the previously installed sources to ensure proper and seamless performance. The long half-life (~433 years) of ^{241}AmBe would provide an almost constant level of neutron flux from the source over the lifetime of the equipment (~20 years). The FMFM would be calibrated once after the ^{241}AmBe source installation, practically eliminating any interference on facility operation for the lifetime of the equipment. The standard size U.S. ^{241}AmBe source (3 Ci in a 0.75-in.-diam, 2-in.-long double-sealed stainless steel capsule) fits into the present FMFM moderator source housing. This source provides ~6.6×10^6 neutron/s, which is equivalent to the neutron output of the ^{252}Cf source, with an average neutron energy of ~4 MeV, a factor of two higher than ^{252}Cf neutrons. Experiments to characterize ^{241}AmBe sources were performed on the FMFM Test Facility, using a ^{3}He-neutron counter and a fission chamber. Thermal neutron production, which is one of the key FMFM performance parameters, and dose rate were evaluated. These experiments are designed to provide data to assess the possible use of ^{241}AmBe for the FMFM. The assessment caveats are that the FMFM performance needs to be maintained and the that facility dose rate requirement must be met (i.e., maximum of 0.3 mrem/h at 1 m from the equipment) without significantly altering the FMFM design. The experimental results on the performance comparison obtained from the ^{252}Cf and ^{241}AmBe sources as well as the overall assessments are presented.

1. INTRODUCTION

The fissile mass flow monitor (FMFM) flow rate measurement principle relies on producing delayed gamma rays emitted from fission fragments carried by UF_6 flow. The FMFM uses thermalized isotopic ^{252}Cf neutron sources for the fission activation of the UF_6 gas stream. The ^{252}Cf neutron sources are placed in an annular sleeve filled with moderator material (made of high-density polyethylene) that surrounds the pipe, as illustrated in Fig. 1. The induced fissions are time-modulated using a neutron-absorbing shutter to create a time signature in the UF_6 gas flow. To satisfy the facility dose rate requirement (i.e., maximum of 0.3 mrem/h at 1 m from the equipment), the FMFM source moderator assembly is surrounded with a lithiated polyethylene shielding, as shown in Fig. 2, and the whole assembly is called the FMFM source modulator (SM).

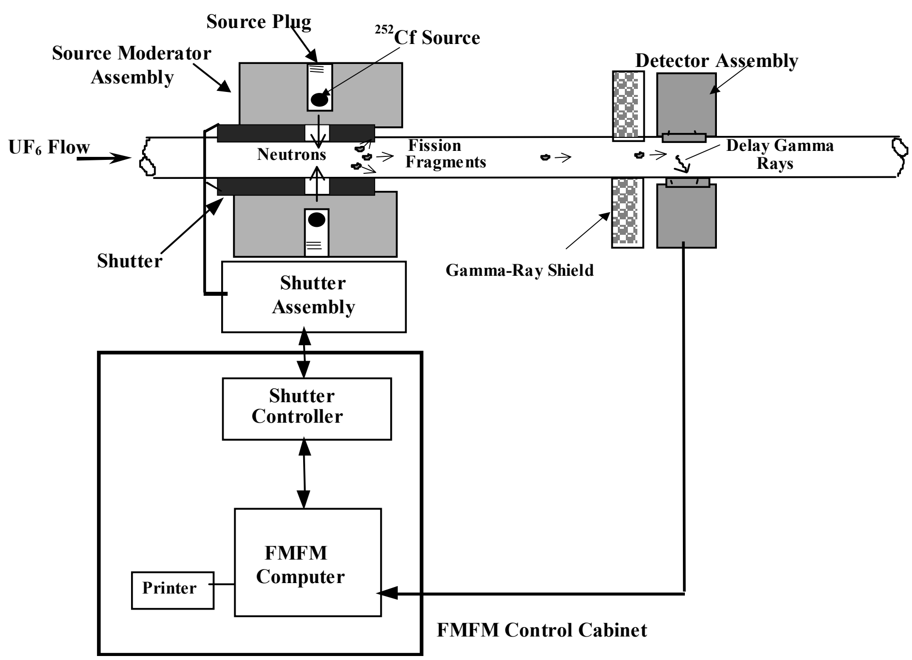

Fig. 1. The Fissile Mass Flow Monitor (FMFM) operational principle and the major components.

A gamma ray detector, located downstream of the sources, measures delayed gamma rays emitted by the resulting fission fragments. The FMFM then determines the fissile mass flow rate from the time-correlated measurement between the SM and the detector signal that provides the

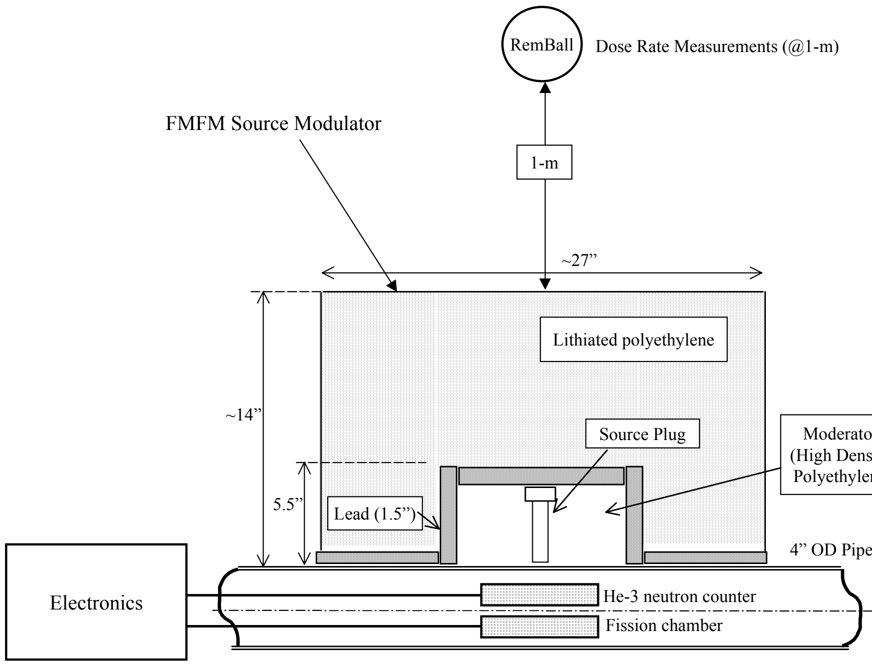

Fig. 2. Details of the FMFM source modulator assembly and instrumentation used for the experiments. (For simplicity, the shutter is not shown.)

UF_6 velocity; the signal amplitude is related to the ^{235}U concentration in the UF_6. The details of the FMFM models employed to predict the detector response are discussed in an earlier publication [1].

The ^{252}Cf source is an isotopic spontaneous fission neutron source that loses its excess energy by alpha emission, which is the dominant decay mechanism (half-life ~2.7 years) over the spontaneous fission process (half-life ~85 years). The ^{252}Cf neutron source output is estimated from the spontaneous fission decay rate, which is ~2.34×10^6 neutron/s per microgram of the sample. The ^{252}Cf neutron source is produced with a very small amount of active element and thus has a small size. This feature makes ^{252}Cf feasible for effective use in the FMFM. The ^{252}Cf neutron energy spectrum extends to ~10 MeV with average neutron energy of ~2 MeV. The FMFM uses a total of four ^{252}Cf sources. Each source contains 3 µg (or ~1.6 millicuries) and provides the neutron output needed for the optimum performance (~6.6×10^6 neutron/s) [1]. The ^{252}Cf sources are made of a double-walled stainless steel encapsulation with a diameter of 0.37 in. and a length of 0.77 in. [2]. The ^{252}Cf sources are placed into the FMFM source moderator with 90^{o} angular separation in special source plugs made of polyethylene (see Fig. 1). The present sources are replaced about every 2 years due to their relatively short half-life (~2.65 years) before the FMFM thermal neutron production performance degrades (that is, when ^{235}U fission production in the UF_6 flow drops to ~60% of the initial value obtained when the ^{252}Cf sources are fresh). The ^{252}Cf source replacement is a costly process, not only for the source fabrication, but also because of the associated cost of transportation, installation, and storage. In addition, the sources are calibrated with the previously installed sources to ensure proper and seamless performance. Moreover, the source replacement activity interferes with the facility operation. In this paper, results of assessment studies performed on the FMFM for possible use of alpha-initiated radioisotopic $^{241}AmBe$ sealed neutron sources (half-life ~433 years) are presented.

2. ALPHA-INITIATED RADIOISOTOPIC SEALED-NEUTRON SOURCES

The main advantages of using alpha-initiated radioisotope neutron sources are (1) physically small size and rugged construction (i.e., a double-walled stainless steel capsule), (2) neutron flux stability, and (3) long useful lifetime (more than 20 years) [3]. These isotopic (α, n) neutron sources are constructed as an intimate mixture (generally in oxide powder form) of an alpha-emitting radioisotope and a target material such as beryllium (see Table 1).

Table 1. Typical long-lived radioisotopic sealed-neutron sources and their characteristics

Source type	Half-life (years)	Source mass for output of ~6.6×10^6 neutron/s (g)	Activity (Ci)
$^{244}CmBe$	18	0.02	1.7
$^{238}PuBe$	87	0.13	2.2
$^{241}AmBe$	433	0.87	2.9

The energetic neutrons are generated following an interaction between the alpha particle and the target material's nucleus and possess energies up to 10 MeV, with an average value of ~4 MeV. Neutrons are produced in typical $^{238}PuBe$, $^{241}AmBe$, and $^{244}CmBe$ radioisotopic sources through the following reaction:

- $^{4}\alpha_{2}$ + $^{9}Be_{4}$ (target) $\rightarrow$ $^{12}C_{6}$ + neutron + 5.71 MeV.
- Alpha emitters: ^{238}Pu, ^{239}Pu, ^{241}Am, and ^{244}Cm.

According to Table 1, $^{238}PuBe$ and $^{241}AmBe$ could provide the necessary neutrons (~6×10^{6} neutron/s) for the FMFM for at least 20 years with a realistic source size (source mass <1 g. These alpha emitters are easily shielded and do not emit penetrating gamma radiation (~60 keV is the dominant gamma ray for ^{241}Am) except for the ~4.4-MeV gamma ray resulting from the deexcitation of ^{12}C. Currently, no U.S. manufacturer fabricates a $^{238}PuBe$ sealed neutron source; however, $^{241}AmBe$ sealed neutron sources are readily available. Therefore, a standard size U.S. $^{241}AmBe$ source (3 Ci in a 0.75-in.-diam, 2-in.-long stainless steel double-sealed-capsule) [4] was selected for the FMFM radioisotopic source assessment studies. ^{241}Am is an alpha emitter and presents an internal health hazard in its loose (^{241}Am oxide powder) form. However, in a $^{241}AmBe$ source, the ^{241}Am oxide is sealed in a double-walled certified sealed source capsule [5] with stainless steel casing and cannot leak out because it is pressed into a solid pellet and is bonded to the casing wall. Furthermore, ~5-MeV alphas have a range of a few centimeters in air [3] and they can easily be stopped. According to the U.S. source manufacturer, many working $^{241}AmBe$ sealed sources have been used in oil fields for more than 30 years without any failure [4]. This standard size source has the following properties:

- Fits into the FMFM moderator source plug housing (see Figs. 2 and 3).
- Can provide ~6.6×10^{6} neutron/s, which is equivalent to the neutron output of present ~3-µg FMFM ^{252}Cf source.
- Average neutron energy is ~4 MeV, a factor of two higher than that of ^{252}Cf neutrons, due to the "hard" neutron energy spectrum.
- Recommended usage time is a minimum of 20 years.

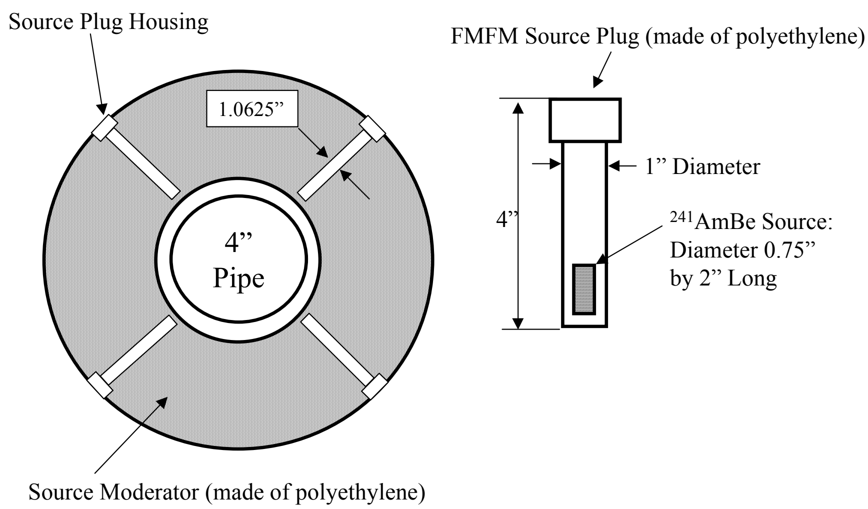

Fig. 3. Details of the FMFM source moderator and the source plug.

3. OBJECTIVE AND EXPERIMENTS

The main objective of this study is to characterize, demonstrate, and assess the use of ^{241}AmBe isotopic sealed neutron sources as alternative long-lived sources (~433 years) to the present short-lived FMFM ^{252}Cf sources without degrading the performance of the FMFM. A set of experiments designed to meet the objective of this study were conducted at the Oak Ridge National Laboratory (ORNL) FMFM Test Facility with ^{241}AmBe sources installed on a 4-in. pipe (see Fig. 4).

The following measurements were carried out:

- The dose rate at 1 m from the FMFM surface (see Fig. 2) with the FMFM shutter closed: the neutron dose rate was measured with a commercially available RemBall (Eberline), and a MicroRem (BICRON) was used for the gamma ray dose rate measurements.
- The thermal neutron production rate inside the 4-in. pipe resulting from the external neutron source: (1) a ^{3}He-neutron counter, which is sensitive to the presence of thermal neutrons, was used (see Fig. 2). (2) A fission chamber measured the fission production rate resulting from the presence of ^{235}U located inside the chamber wall when exposed to thermal neutrons. The chamber was placed inside the pipe together with the ^{3}He counter under the source (see Fig. 2).

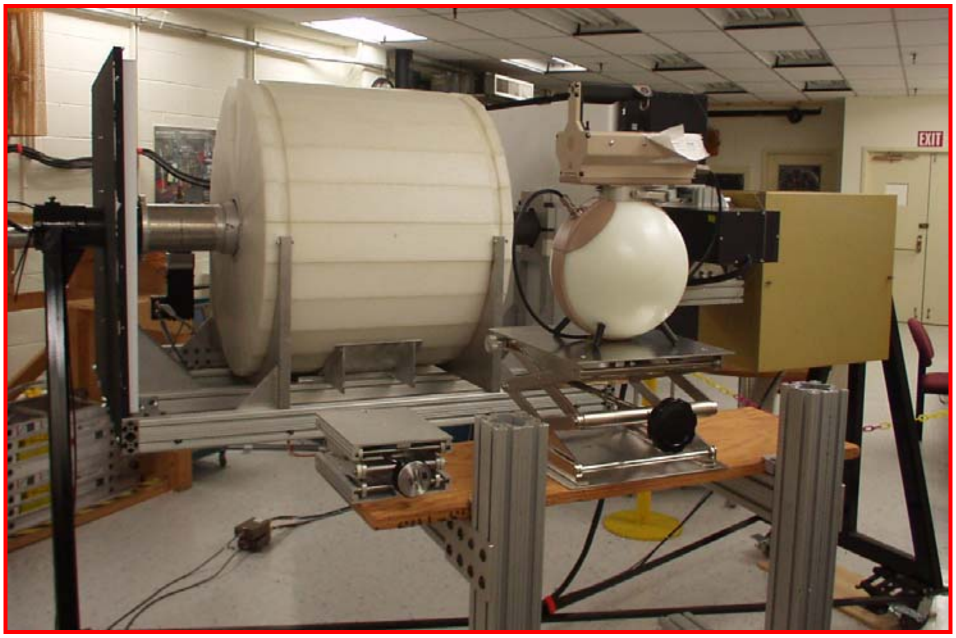

Fig. 4. The ORNL FMFM Test Facility source modulator and the experimental arrangement.

These experiments provided data to evaluate and assess a possible use of ^{241}AmBe sources in the next-generation FMFM. The main caveat used for the assessment of the results was to maintain the FMFM performance obtained with the ^{252}Cf sources. This assessment was accomplished by comparing the results of experiments carried out with ^{241}AmBe and ^{252}Cf sources. The experiments were performed by having the same source neutron output of a nominal ~6.6×10^6 neutron/s provided either from the ^{252}Cf source, which is about 3 µg, or the ^{241}AmBe neutron source, which is about 2.9 Ci in activity. The comparison measurements of the ^{252}Cf and the ^{241}AmBe sources for their dose rates at 1 m from the FMFM were carried out for the following source locations (see Fig. 5):

1. The source of interest was placed in the location indicated by Top/Front on the SM, and the corresponding dose rate, denoted by D_{front}, was determined.
2. The source of interest was placed in the location indicated by Top/Back on the SM, and the corresponding dose rate, denoted by D_{back}, was determined.

The total FMFM dose rate, which is an equivalent to having four sources placed symmetrically around the source moderator (see Fig. 3), was then obtained by $D_{total} = 2 \times (D_{front} + D_{back})$. In the

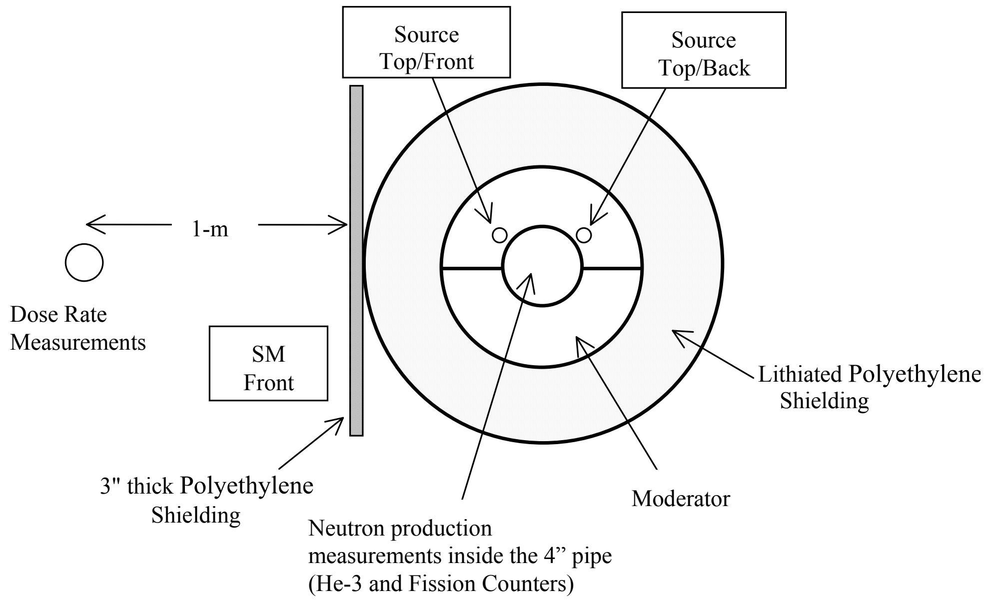

Fig. 5. Cross-sectional view of the FMFM source modulator showing the arrangement of the source installation locations.

case of the ^{241}AmBe source, the dose rate measurements were carried out both with and without an additional 3 in. of polyethylene shielding placed at the front of the SM, as shown in Fig. 5.

4. RESULTS

The results obtained from the neutron and the gamma-ray dose rate measurements for the ^{252}Cf and ^{241}AmBe sources are summarized in Table 2. The total dose rates are provided for measurements taken both with and without a 3-in.-thick polyethylene shielding panel placed in front of the SM, as shown in Fig. 5. The measured ^{241}AmBe source total dose rate, ~0.63 mrem/h at 1 m from the FMFM, is higher than that of ^{252}Cf by about a factor of 2.6. With the placement of the additional polyethylene shielding, this factor reduced to about 1.57, a significant drop in the neutron dose rate (see Table 2). The thermal neutron production rate (which is related to the fission production rate in the UF_6 gas inside the pipe) resulting from the moderated ^{241}AmBe source neutrons, normalized to the ^{252}Cf source, is about 0.75 (see Table 3). This averaged value is obtained from the measurements made by the ^{3}He neutron counter (~0.8) and the fission chamber (~0.7) placed inside the pipe, as illustrated in Fig. 2. The thermal neutron production rate inside the pipe is also determined with Monte Carlo modeling simulations [6] to estimate the

Table 2. Measured dose rate results for ^{252}Cf (~3-μg) and ^{241}AmBe (2.91-Ci) sources[a]

Emission	Dose rates (mrem/h)			^{241}AmBe to ^{252}Cf ratios	
	^{252}Cf	^{241}AmBe	^{241}AmBe, shielded[b]	Unshielded	Shielded[b]
Neutron	0.1975	0.5856	0.3221	2.965	1.631
Gamma	0.0307	0.0448	0.0364	1.461	1.187
Total	0.2282	0.6304	0.3585	2.763	1.571

[a]Dose rate at 1 m in front of the source modulator; four sources installed; 6.6 × 10^6 neutron/s per source.
[b]SM shielded with 3 in. of polyethylene.

Table 3. ^{241}AmBe dose rates and the normalized thermal neutron production at various source activities[a]

^{241}AmBe source strength		Equivalent ^{252}Cf source strength (μg)	Neutron production normalized to ^{252}Cf	Total dose rate (mrem/h)[b]
× 10^6 neutron/s	Ci			
6.6	2.91	2.9	0.75	0.412
6	2.65	2.6	0.68	0.375
5.5	2.43	2.4	0.63	0.344
5	2.20	2.2	0.57	0.312
4.5	1.98	2.0	0.51	0.281
4	1.76	1.7	0.45	0.250

[a]Front of SM covered with an additional 3 in. of polyethylene shielding.
[b]Instrument error: 15%; measured at 1 m in front of SM; SM loaded with four sources.

effectiveness of the ^{241}AmBe source relative to that of the ^{252}Cf source used at the nominal value for the FMFM. The fission production rate in the UF_6 gas induced by the ^{241}AmBe neutron source was predicted to be about 72% of that induced by the ^{252}Cf source. This prediction is consistent with the measurement results. In addition, as also predicted, this value is not very sensitive to the radial location of the source position inside the FMFM moderator (see Fig. 3).

The results given in Table 2 are extended in Table 3 to include the normalized thermal neutron production rate as well as the measured dose rate upper bounds obtained from consideration of instrument error (~15%). The results are given as a function of the quantity of ^{241}AmBe per source activity, starting from the nominal value of ~2.9 Ci (which produces the ^{252}Cf equivalent neutron output of ~6.6 × 10^6 neutron/s) down to 1.76 Ci, which is the source value at which the total FMFM dose rate becomes well below the acceptable facility level of 0.3 mrem/h.

For example, in the case of the 1.76-Ci ^{241}AmBe source, the total dose rate is 0.25 mrem/h, while the thermal neutron production rate is ~45% compared with the nominal ^{252}Cf source production rate. Even though the facility dose rate requirement is satisfied in this case, the FMFM performance is significantly degraded (by more than 50%). This FMFM performance degradation results in a lower detector signal amplitude, S_d, and in turn, longer signal convergence times, $t_c \sim 1/S_d^2$, are needed to obtain statistically acceptable results.

5. ASSESSMENT OF RESULTS

The results given in Table 3 were further studied to match the performance of the ^{241}AmBe source with that of the ^{252}Cf source (namely the thermal neutron production rate must be kept at the same value). This was achieved by increasing the ^{241}AmBe source activity by about 25%. In Table 4, summaries are given of the results of the ^{241}AmBe thermal neutron production rate normalized to that of ^{252}Cf, the total dose rate with the additional 3 in. of polyethylene at various reduced values of ^{241}AmBe source activity, together with the equivalent ^{252}Cf source masses, until the facility dose rate is satisfied (i.e., below 0.3 mrem/h at 1 m from the FMFM).

As seen in Table 4, when the ^{241}AmBe source activity reaches 8 Ci, which is equivalent to 6 µg of ^{252}Cf, the FMFM performance is 50% that of the ^{252}Cf, and the total dose rate upper bound is 0.28 mrem/h. In this case, the detector signal is reduced by a factor of about two, resulting in signal convergence times that are longer by a factor of about four. This is significant performance degradation because the FMFM measurement results are often needed from the shortest measurement times possible.

Table 4. ^{241}AmBe source assessment results compared with ^{252}Cf performance[a]

^{252}Cf (µg)	^{252}Cf neutron production relative to 12 µg of ^{252}Cf	Total ^{241}AmBe activity (Ci)	Maximum total dose rate (mrem/h)
12	1.000	16.000	0.567
10	0.833	13.333	0.472
8	0.667	10.667	0.378
6	0.500	8.000	0.283
5	0.417	6.667	0.236

[a]SM covered with an additional 3 in. of polyethylene shielding; measurements taken at 1 m in front of the SM.

6. SUMMARY AND CONCLUSIONS

The experimental results on the performance comparison obtained from the ^{252}Cf and ^{241}AmBe sources carried out on the ORNL FMFM Test Facility can be summarized as follows: (1) The ^{241}AmBe source thermal neutron production rate performance is ~25% lower than that of the ^{252}Cf source. (2) The dose rate is ~35% higher (with additional shielding) due to the presence of high-energy ^{241}AmBe neutrons. If the frequent FMFM ^{252}Cf source replacement is not desirable, then implementation of the ^{241}AmBe source is possible provided that (1) the FMFM is modified to accommodate additional neutron shielding and (2) a larger FMFM design is considered for creating a higher fission production rate to compensate for the 25% degradation. These two suggested FMFM modifications should be studied as trade-off assessments between the desired signal convergence times and a realistic FMFM equipment design size for determining the proper source activity. The next-generation FMFM with a successful ^{241}AmBe source implementation can have the following benefits:

- There is no need to replace the sources for the lifetime of the equipment; therefore, there is no interference with facility operation.
- The FMFM performance continuity is achieved through time-independent neutron flux.
- The FMFM detector signal remains constant (for the same operating conditions), leading to constant measurement-convergence times.
- Detecting any unexpected changes in the facility operation is easy.
- It eliminates concerns about whether replaced sources are the same as the original ones.
- It reduces the cost associated with the source replacement.

REFERENCES

1. J. March-Leuba, J. K. Mattingly, J. A. Mullens, et al., *38th Annual INMM Meeting*, Phoenix, Arizona, July 20, 1997.

2. Frontier Technology Corporation, 1641 Burnett Drive, Xenia, OH 45385.

3. G. F. Knoll, *Radiation Detection and Measurements*, John Wiley & Sons, New York, 1989.

4. Gammatron Inc., P.O. Box 266677, Houston, TX 77207.

5. Meets requirements for special form as defined in DOT Title 49 (173.403[z]).

6. J. K. Mattingly, ORNL, private communication.

INTERNAL DISTRIBUTION

1. J. A. March-Leuba
2-6. D. H. Powell
7-11. T. Uckan
12. J. D. White
13. Central Research Library
14. ORNL Laboratory Records – RC
15. ORNL Laboratory Records - OSTI

EXTERNAL DISTRIBUTION

16. Guy Armantrout, Lawrence Livermore National Laboratory, U. S. Department of Energy, NA-23/Germantown Building, 1000 Independence Avenue, S. W., Washington, DC 20585
17. Janie Benton, U. S. Department of Energy, NA-23/Germantown Building, 1000 Independence Avenue, S.W., Washington, DC 20585-1290
18. Dianna Blair, Sandia National Laboratory, International Programs, 10600 Research Road, Albuquerque, NM 87123
19. Cynthia Boggs, Argonne National Laboratory c/o 270 Complex, 19901 Germantown Road, Germantown, MD 20874
20. Julie Bremser, Los Alamos National Laboratory, MS J562, Bikini Atoll Rd., SM 30, Los Alamos, New Mexico 87545
21. Donald Close, Los Alamos National Laboratory, MS J562, Bikini Atoll Rd., SM 30, Los Alamos, New Mexico 87545
22. David Dougherty, Brookhaven National Laboratory, U. S. Department of Energy, NA-241, 1000 Independence Avenue, S.W., Washington, DC 20585
23. Melvin Feather, II, SAIC, 20201 Century Blvd., Ste. 300, Germantown, MD 20874
24. Joseph Glaser, U. S. Department of Energy, NA-232, 1000 Independence Avenue, S.W., Washington, DC 20585
25. Kenneth Lewis, DOE/NBL, New Brunswick Laboratory, 9800 S. Cass Avenue, Argonne, IL 60439
26. Edward Mastal, U. S. Department of Energy, NA-232, 1000 Independence Avenue, S.W., Washington, DC 20585
27. Calvin Moss, Los Alamos National Laboratory, MS J562, Bikini Atoll Rd., SM 30, Los Alamos, New Mexico 87545
28. Radoslav Radev, Lawrence Livermore National Laboratory, P. O. Box 808, Livermore, CA 94551
29. Robert Richmond, Bechtel Nevada, P. O. Box 380, Suitland, MD 20752
30. Kurt Siemon, Jr., U. S. Department of Energy, DOE/NA-241, 1000 Independence Ave., S.W., Washington, DC 20585
31. David Wall, U. S. Department of Energy, NNSA, Y-12 Site Office, MS 8009, 200 Administration Road, Oak Ridge, TN 37831