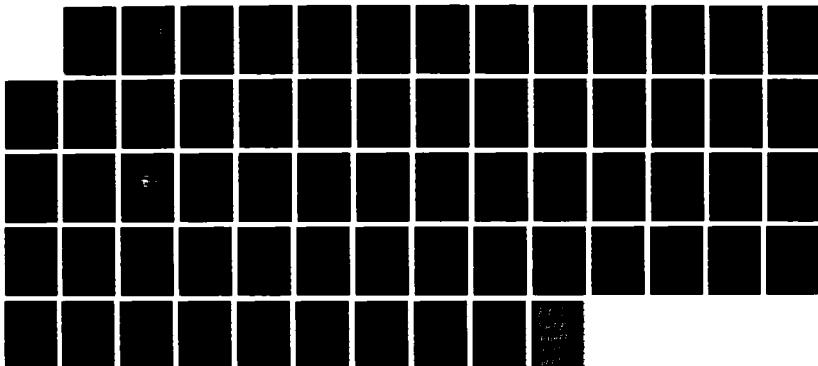
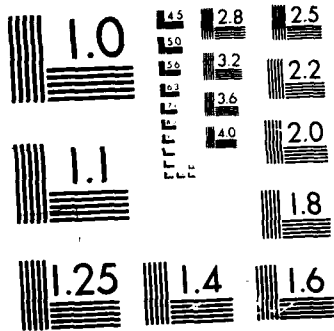


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DETERMINATION OF PU-BE SOURCE STRENGTH
 BY MANGANESE ACTIVATION

THESIS

Paul F. Whitworth Jr.
 Captain, USA

AFIT/GNE/ENP/88M-10



DEPARTMENT OF THE AIR FORCE
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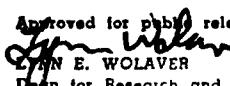
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**DETERMINATION OF PU-BE SOURCE STRENGTH
BY MANGANESE ACTIVATION**

THESIS

Paul F. Whitworth Jr.
Captain, USA

AFIT/GNE/ENP/88M-10

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DETERMINATION OF PLUTONIUM-BERYLLIUM SOURCE
STRENGTH BY MANGANESE ACTIVATION

THESIS

Presented to the Faculty of the School of Engineering
of the Air Force Institute of Technology

Air University

In Partial Fulfillment of the
Requirements for the Degree of
Master of Science in Nuclear Engineering

Paul Fletcher Whitworth Jr. BS,

Captain, USA

March 1988

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Preface


This thesis describes my efforts to determine the total neutron output of a Pu-Be neutron source. The objectives of this study were: 1) to examine the method and factors involved, 2) accurately determine the correction factors used, 3) determine the neutron source strength of AFIT's neutron source, and 4) provide a technique and correction factors that others could use. This thesis will summarize past work in this area, describe the procedure, and present the results, an analysis of the results, conclusions, and recommendations.

I wish to thank Dr. Denis Beller, my thesis advisor, for his continued help and guidance, Dr. George John for his guidance through many experiments, and Mike Archuleta for the time he invested in this thesis. I would also like to thank my other classmates, for their help and support. Finally, I am especially grateful to my wife, Ruth, for her patience and support, and to my son, Stephen, who reminded me that even at its worst, it's not that bad.



ABSTRACT

In this study the manganese-sulfate-bath technique was used to determine the total neutron output of a Plutonium-Beryllium neutron source. Activation trials were conducted using 4 different concentrations of manganese sulfate and 3 different size containers. Correction factors for source absorption, non-leakage, and resonance absorption were calculated using data generated by *MORSE*, a Monte Carlo neutron transport code. Values for these factors were calculated for manganese sulfate concentrations between 5.56% and 36.00%, and volumes from 17.2 to 160 liters in cylindrical containers. A source strength of $1.07 \times 10^7 \pm 3\%$ neutrons per second was found by using experimentally measured manganese activity in conjunction with the computer-predicted correction factors.



10 to 110

I. INTRODUCTION

Plutonium-Beryllium neutron sources do not maintain a constant source strength over long periods of time (years). The Air Force Institute of Technology possesses a neutron source, M-1170, prepared by Mound Laboratory in 1962. Metzler's preliminary studies of the total neutron output of this source in 1986 identified the correction factors and the sources of uncertainty. However, he was unable to obtain accurate results because he lacked the time and analytical tools to calculate accurately some of the correction factors needed to determine the total neutron output (13). This study, examines the experimental methods and correction factors, determines the total neutron output of the source, and provides a set of correction factors for others to use when calibrating the neutron source.

Background

Neutrons are produced in this source when α particles emitted by the decay of ^{239}Pu interact with ^9Be by the reaction $^9\text{Be}(\alpha, n)^{12}\text{C}$. Some of these neutrons, especially when thermalized by surrounding materials, will cause other ^{239}Pu nuclei to fission, producing more neutrons. When the source was constructed, some ^{241}Pu was present in the ^{239}Pu . Plutonium-241 (half-life 14.4 years) decays by beta emission to produce Americium-241 (half-life 432 years). This increases the number of neutrons since the ^{241}Am concentration increases with time and emits α particles as it decays. As the concentration of ^{241}Pu increases, the neutron output of the source increases, reaching a peak in about 70 years. The product specification that accompanied the source listed the fraction of ^{241}Pu present at the time of manufacture and supplied an equation to determine the source strength at later dates. This equation was used as a check for the experimentally determined values.

Review of the Literature

Metzler used an aqueous manganese sulfate solution, 5.56% $MnSO_4$ by weight, in a spherical, 22 liter, pyrex flask to perform his activation experiment. He attempted to extrapolate values for factors that he could not measure directly from calculations made by other researchers. The differences in the sources used by the other researchers and M-1170 resulted in inaccuracies in several of the correction factors. Also, pyrex contains approximately 15% B_2O_3 by weight. Approximately 20% of naturally occurring boron exists as ^{10}B , which has a 3838 barn thermal neutron cross section. The boron in the pyrex flask probably introduced some error into the leakage measurements (13). Metzler determined the neutron source strength to be $(7.8 \pm 2.6) \times 10^6$ neutrons per second. This value was 28% lower than the calculated value, and had a systematic uncertainty of 33%.

Other researchers have used the manganese-bath technique to calibrate neutron sources and to determine thermal neutron cross-sections of several nuclides. Geiger and Whyte used a 30.31% $MnSO_4$ solution to determine the source strength of a $Ra-Be$ neutron source. They used a cylindrical steel tank, 85 cm in diameter to contain the bath. To measure the activity of the ^{56}Mn , they inserted a "commercial Geiger-Muller tube" into the center of the bath. The neutron output was determined within 2% of the value determined by the National Bureau of Standards (7).

Hanna and Runnalls used the activation of ^{55}Mn in an aqueous $MnSO_4$ solution to determine the source strengths of 4 actinide-beryllium sources. They used a 31.4% $MnSO_4$ solution in a steel tank 24 inches in diameter, filled to 24 inches in height. They used a sodium Iodide scintillation counter, 2 inches in diameter and 1 inch thick, lowered into the bath, to determine the ^{56}Mn activity. The

source strength values they obtained using the manganese activation method were within 2% of the values obtained by measuring the source strength with a BF_3 counter, calibrated with a Ra-Be standard from the National Bureau of Standards (9).

Hwang and Choi used a circulated manganese bath in a circular steel container, 125 cm in diameter to develop a system to calibrate neutron sources at the Korea Standards Research Institute. Their system used circulating pumps to mix the bath and route a portion of the liquid through a Marinelli beaker where it was counted by a sodium iodide detector. Calibration checks using an Am-Be and a Cf-252 as sources of neutrons produced results with less than 1% error (10).

Arbildo used the manganese bath technique to determine thermal neutron cross-sections for boron, manganese, and sulfur. His method was based on the comparison of ^{56}Mn activity produced by a Sb-Be neutron source in a manganese sulfate bath. By controlling the proportions of the elements in the bath he was able to accurately determine the cross-section ratios of manganese, boron, and sulfur to hydrogen, whose thermal cross-section is accurately known. His work produced more accurate estimates for the thermal cross-sections for these three nuclides (1).

Problem

The problem investigated in this study was the determination of total neutron output of a Pu-Be neutron source, specifically, serial number M-1170 made by Mound Laboratory. The method used for the output determination was to activate several concentrations of manganese sulfate in aqueous solution, and to relate the activity of the ^{56}Mn to the source strength through the use of several correction factors. The correction factors were obtained by implementing a computer code, called *MORSE*, obtained from Oak Ridge National Laboratory, which

was used to create a computer model of the experiments. The correction factors were calculated from the results obtained from the computer model. The details of these calculations are addressed in *Procedure*.

Scope

This study was primarily concerned with determining the total neutron output of the Pu-Be source by the activation of ^{55}Mn . This determination required measurement of activities of ^{56}Mn and the development of an analytical method to calculate factors such as system leakage, resonance absorption, and source absorption. The accurate determination of these correction factors was the primary objective of this study. The goal of the study was to use the correction factors to accurately estimate the total neutron output of the source with a precision of 3 % or better. The study was limited to manganese sulfate baths in cylindrical containers with diameters ranging from 27 to 54.4 cm. and height 1.22 times the diameter. Concentrations ranging from 5.56% to 36% MnSO_4 by mass were examined. The energy distribution of the neutrons emitted by the source was not measured, but was examined in the literature search and from the *MORSE* computations. Values for the correction factors obtained from the computer model were compared to values obtained by experimental methods and to values obtained by other researchers.

Assumptions

The major assumption in this study was that all thermal neutrons that did not leak from the system were absorbed by manganese, sulfur, or hydrogen. Absorption by other nuclides was considered insignificant. The primary energy spectrum of the neutron source was assumed to be the same as the spectrum

reported by Kumar and Nagarajan (11: 177) for similar Pu-Be neutron sources. The fission spectrum for ^{239}Pu presented by Bartine et al., was assumed to be accurate for this study.

Organization

This report is presented in 5 major sections: I) the introduction; II) theory; III) procedure; IV) results and discussion of results; and V) conclusions and recommendations. In addition, the Appendix includes a list of equipment used in this study and the number densities of the MnSO_4 solutions used in the computer calculations.

II. THEORY

When a neutron source is introduced into a manganese sulfate solution, the fast neutrons emitted by the source are thermalized by the water and absorbed by the manganese, hydrogen, and sulfur in the solution (some neutrons are absorbed by the oxygen, but the thermal neutron cross-section of oxygen is five to six orders of magnitude below the others (16: 8-1) and is negligible. If the solution is irradiated for a very long time (approaching infinity), the system reaches equilibrium, where the decay rate and production rate of ^{56}Mn are equal. The activity of the ^{56}Mn can then be related to the output of the neutron source by:

$$Q = \frac{A_s}{MAF} \quad (2.1)$$

where Q is the neutron source strength, A_s is the saturation activity of ^{56}Mn , and MAF (*M*anganese *A*bsorption *F*raction), the fraction of the absorbed neutrons that are absorbed by manganese. The MAF is determined by:

$$MAF = \frac{(N\sigma)_{Mn}}{(N\sigma)_{Mn} + (N\sigma)_S + (N\sigma)_H} \quad (2.2)$$

where N and σ are the number and microscopic cross-section for each type atom.

Eq 2.2 is true however, only if the following occurs:

- 1) No neutrons escape;
- 2) No neutrons are absorbed before they are thermalized;
- 3) No neutrons are absorbed by the source; and
- 4) Irradiation occurs long enough for equilibrium to be established (infinite irradiation time).

Factors are introduced to correct for leakage, resonance absorption, source absorption, and finite irradiation time. These factors are briefly described below and will be discussed in more detail later.

LEAKAGE: (nlf) Since the bath is not infinite, some fraction of the source neutrons will escape or leak out. This is the leakage fraction, lf . The fraction of neutrons that do not leak and are available for absorption, the non-leakage fraction, is then:

$$nlf = (1 - lf) \quad (2.3)$$

RESONANCE ABSORPTION: (p) Not all of the source neutrons will become thermalized. Two nuclides in the bath have significant cross-sections for high energy neutrons (above 2 Mev), oxygen and sulfur. Absorption of neutrons by nuclides of these elements reduces the number of neutrons available for absorption by manganese (6: 259). ^{55}Mn does not have a significant high-energy absorption cross-section. The fraction of source neutrons that are not captured at high energies is the resonance escape fraction, p .

SOURCE ABSORPTION: (sa) Some neutrons will be reflected back into the Pu-Be source. The neutrons entering the source can be absorbed by the case material of the source, reducing the number of neutrons available for absorption, or they can be absorbed by ^{239}Pu , which has a 742 barn thermal neutron cross-section, and produce additional fission neutrons. The fission of ^{239}Pu uses 1 neutron and produces an average of 2.87 neutrons for an average net increase of 1.87 neutrons per fission (12: 68). Since the output of the source may be increased by the neutrons reflected by the bath, the Source Absorption factor may be greater than one.

FINITE IRRADIATION TIME: The activity of ^{56}Mn , after an irradiation time t_{irrad} is related to the production rate of ^{56}Mn , R by:

$$R = \frac{A(t)}{(1 - \exp(-\lambda t_{\text{irrad}}))} \quad (2.4)$$

where λ is the decay constant for ^{56}Mn , 0.2688 hr^{-1} (3: 25) When the activity of the ^{56}Mn at source removal (time t) is determined, the activity of the manganese after infinite irradiation can be calculated.

When these correction factors are included, Equation 2.1 becomes:

$$Q = \frac{A(t)}{MAF \cdot p \cdot nlf \cdot sa \cdot (1 - \exp(-\lambda t_{\text{irrad}}))} \quad (2.5)$$

where $A(t)$ is the activity at the time of source removal, t_{irrad} is the irradiation time, and the other factors are as indicated earlier. When the activity of the ^{56}Mn is measured, the value of $A(t)$ can be calculated from the net count in a counting time t_c by:

$$A(t) = \frac{\lambda C}{E_p \cdot bf \cdot \exp(-\lambda t_r) \cdot (1 - \exp(-\lambda t_c))} \quad (2.6)$$

where E_p is the absolute peak efficiency of the counting system for 847 keV gammas, bf is the branching factor for 847 keV gammas, C is the number of net counts in the 847 keV full energy peak, and t_r is the amount of time between the removal of the source and the beginning of the measurement. The value for $A(t)$ from equation 2.6 is then used in equation 2.5 to find the total neutron output, Q , of the source in neutrons/unit time (ie. neutrons/second).

Computer Model Determination of Correction Factors

Several of the correction factors necessary for the source strength determination were difficult, if not impossible to measure directly. Therefore, a computer code was used to model the experiments, and to produce data that could be used to calculate the correction factors. The computer code chosen for the study was MORSE, a multi-group, neutron and gamma transport code, using the Monte Carlo method for radiation transport calculation (6).

The Monte Carlo method uses the random movements and interactions of a large number of particles to determine an average behavior of the particles as a whole. The distance a particle travels and the types of interactions that it undergoes are determined by probability density functions and random numbers. When a particle is produced or introduced into a given medium, random numbers are generated that determine the particle's track length and type of interaction. If the interaction is a scatter, random numbers are generated that determine the speed, direction and distance of the particle's new path. The process continues until the particle is absorbed, or passes beyond the edge of the medium. Another particle is then introduced, and the process is repeated until all of the particles used in the calculation have been absorbed or escape. A record is kept of what happens to each particle, and the average particle behavior is determined from this record (4: 348).

In MORSE, particles are not absorbed, but are given an initial, statistical, weight, which can be envisioned as some number of particles. As particles interact, their weight is reduced by the ratio of the scattering cross-section to the total cross-section for that medium and energy group. The movement and interactions of the particles are calculated by the Random Walk module in the MORSE code (6: 4.4-1). The subroutine, BANKR, is the record-keeping portion of the code, and is called from the other modules when some part of a particle his-

tory needs recording. Figure 2-1 shows how the basic structure of MORSE is configured (15: 5-2).

Since particles are not absorbed in MORSE, particle histories are terminated in this study by escape, energy, and Russian roulette. When a particle passes beyond the boundary of the of the region in the calculation, it has escaped, and a new particle is introduced. Escape is the same as leakage from the system. A particle is also terminated if its energy passes below the lower energy bound of the lowest energy group. Russian roulette is used to retire particles whose statistical weight has become too low to have a significant bearing on the estimation of the particle flux (15: 2-10).

Several options were available for flux estimation, and the method of track length estimation was chosen. This method of flux estimation used the total, weighted, particle track length divided by the volume of the detector used to determine an average flux (or fluence) for the detector (5: 50). The volume of the detector was chosen to correspond to the same volume as the manganese sulfate bath. The subroutine that calculated the fluence, *TRKCOL* was modified to calculate the total number of absorptions in the bath.

Since the Monte Carlo method is a statistical transport method, some statistical uncertainty is present in the results of the calculations. Normally, the total number of particles used in a calculation are divided into a given number of "batches." When all of the particles in each of the batches has been retired, the computer code calculates a mean value and a standard deviation for the parameters that are being calculated. Increasing the number of particles in each batch, and the total number of batches that are used (which increases the total number of particles) decreases the statistical uncertainty of the calculations. However, this also increases the amount of computer time required to complete the calculation. In this study, the statistical uncertainty in the values calculated by the code

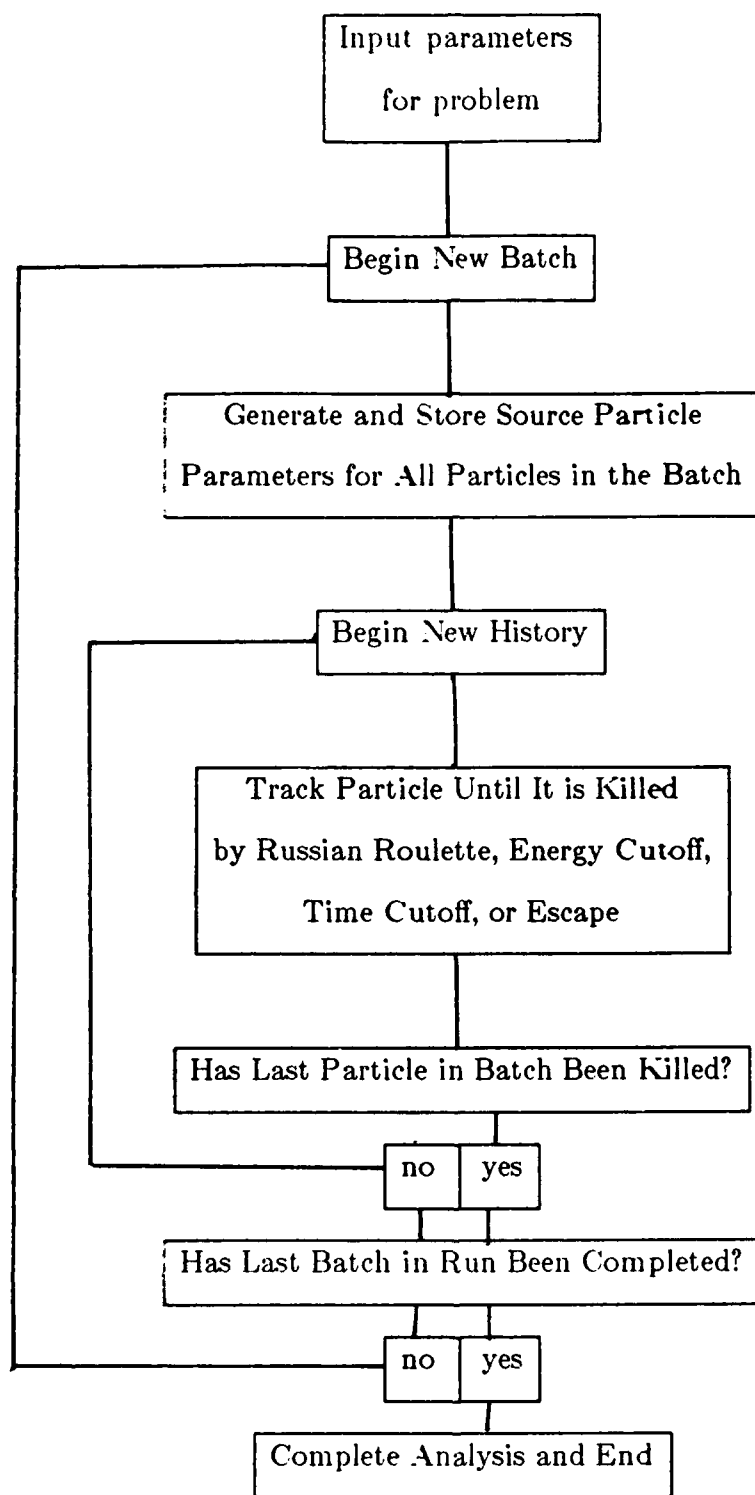


Figure 2-1. The Basic Structure of MORSE

were kept at about 1% by using 50 batches containing 200 neutrons for each trial.
The handling of the statistical uncertainties are discussed in *Procedure*.

III. PROCEDURE

In order to determine the activity of the ^{56}Mn , an experiment was performed to activate several concentrations of aqueous manganese sulfate. A computer model of the experiment was developed to find values for several of the correction factors. This chapter discusses the procedures of the experiment, the development of the computer model, and the calculation of the correction factors.

Experimental Method

Preparation of Bath.

The manganese sulfate solutions were prepared by dissolving monohydrous manganese sulfate ($\text{MnSO}_4 \cdot \text{H}_2\text{O}$) in distilled water. A total of 40 kg of the $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ was dissolved into 60 kg of water in 3 separate batches. The $\text{MnSO}_4 \cdot \text{H}_2\text{O}$, in powder form, was listed by the manufacturer as 99.5% $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ (8). The powder was mixed into the distilled water and stirred with an electric drill. Insoluble impurities were removed by decanting, and samples of the solution were taken to determine an accurate MnSO_4 fraction. This concentration, $(36.00 \pm 0.04)\%$ MnSO_4 , was considered the full concentration level for the experiment. This solution was later diluted to $3/4$, $1/2$, $1/4$, and $1/8$ the original value for portions of the experiment using lower manganese sulfate concentrations. Approximately 21 liters of the $(5.56 \pm 0.01)\%$ MnSO_4 solution used by Metzler were available. This solution had the lowest manganese concentration used in the activation trials.

The actual concentrations of the solutions were determined by evaporating and heating to produce anhydrous MnSO_4 . Measured amounts of the solution samples were placed in porcelain crucibles and put under a heat lamp to evaporate the water. When the majority of the water had evaporated, the crucibles were

then heated in a furnace to approximately 500° C to remove any remaining water and produce anhydrous $MnSO_4$. Concentrations of each bath used for the activation experiments were measured in this manner with an error of not more than 0.2%.

Calibration of Gamma Detector.

This experiment was conducted using a variety of $MnSO_4$ concentrations. Since one of the purposes of the study was to provide a calibration method for most concentrations, the efficiency of the detector measuring the activity of the ^{56}Mn had to be determined for a variety of $MnSO_4$ concentrations. Since the manganese, sulfur, and oxygen in the measurement container (a Modified Marinelli Container, or MMC) could absorb or scatter the 847 keV photons emitted by the ^{56}Mn , the efficiency of the detection geometry varied with the $MnSO_4$ concentration. To account for this change in efficiency, a manganese free or "zero concentration" efficiency was determined, which was modified by a "concentration adjustment" factor.

The detector itself was a 90 cc (active volume), lithium drifted, germanium detector, more commonly called a Ge(Li). Pulse height spectra from the detector were obtained with a Nuclear Data ND-66 multichannel analyzer and ND-680 computer. The ND-680 analyzer contained an efficiency for the measurement geometry, which was made by Leonard (13: 9) in 1984. This was made by placing a liquid, multi-radionuclide standard in a measurement container and calculating an efficiency for the photons that were emitted by the radionuclides in the standard. The ND-680 determined an energy vs. efficiency curve for the MMC geometry. This efficiency was rechecked using a 1986 Amersham standard source prepared in the same manner. This efficiency was used as the "zero concentration" efficiency since the MMC contained no $MnSO_4$.

The concentration adjustment factor was found using a combination of methods. A 0.15 g mass of Mn_2O_3 , in a gelatin capsule, was irradiated at the research reactor at Ohio State University. The activity of the ^{56}Mn was determined by measuring the capsule as a point source, using a known efficiency in the detector. The Mn_2O_3 was then dissolved in an MMC containing the 5.56% solution. The activity of the ^{56}Mn was measured in the MMC geometry, using the zero-concentration efficiency. By dividing the activity in the MMC geometry by the activity in the point geometry, the concentration adjustment factor was determined.

During another trial at the OSU reactor, small polyethylene containers containing approximately 5 ml of the 36.00% and 5.56% solutions were irradiated. These containers had the same dimensions as the standard source vial, and an efficiency was determined for the source in the vial. The activity of the ^{56}Mn in these containers was determined by measuring them in the detector, using the efficiency for the standard source in the vial. The contents of the polyethylene containers were poured into MMC's containing the appropriate solutions, and activities were measured in the MMC geometry. Concentration adjustment factors were again determined.

The concentration adjustment factors for the intermediate concentrations were determined during one of the activation trials. After irradiating the 36.00% solution, measured amounts of the solution were diluted to 18.00, 9.00, and 5.00 percent in MMC's. Uncertainties in the concentrations were kept below 0.05. Activities were measured for each of the concentrations. The actual activity of the concentrated solution was determined by using the concentration adjustment factors from the previous OSU trial. Once the actual activity was found, the concentration adjustment factors were determined for the intermediate concentrations. The 5.00% dilution served as a check, to insure agreement with the concen-

tration adjustment factors calculated using the other methods.

Since the ND-680 calculated the activity of the ^{56}Mn directly, Eq. 2.6 was not required to calculate the activity. However, the concentration adjustment factor affected ϵ_p in Eq. 2.6. The concentration adjustment factor, ca was included in Eq. 2.5 to correct the activity calculated by the ND-680 to the actual activity, forming Eq. 3.1.

$$Q = \frac{A(t)}{MAF \cdot p \cdot nlf \cdot sa \cdot (1 - \exp(-\lambda t_{\text{irrad}})) \cdot ca} \quad (3.1)$$

Manganese Activation Trials.

A total of 13 irradiation trials were conducted using the neutron source and four concentrations of the manganese sulfate bath. Three different sizes of cylindrical polyethylene containers, 17.8, 40, and 140 liters in volume were used. In each trial, the neutron source was placed in the bath and allowed to irradiate the manganese sulfate for 1-2 hours. Figure 3-1 depicts the dimensions of the source (13: 8). After the source was removed, the bath was stirred for 15-20 minutes using an electric hand drill and stirrer. A portion of the solution was then transferred to an MMC and placed in the gamma detector. The gamma detector and ND-680 measured the activity of the ^{56}Mn and calculated the activity at the time the source was removed. The spectra were stored on computer disks in the ND-680. The total activity of the ^{56}Mn in the bath was then calculated by dividing the activity in the MMC by the fraction of the solution mass in the MMC and by the appropriate concentration efficiency adjustment factor.

An experimental non-leakage factor was determined using a BF_3 "long counter," of the type designed by Hanson and McKibben (17: 328).

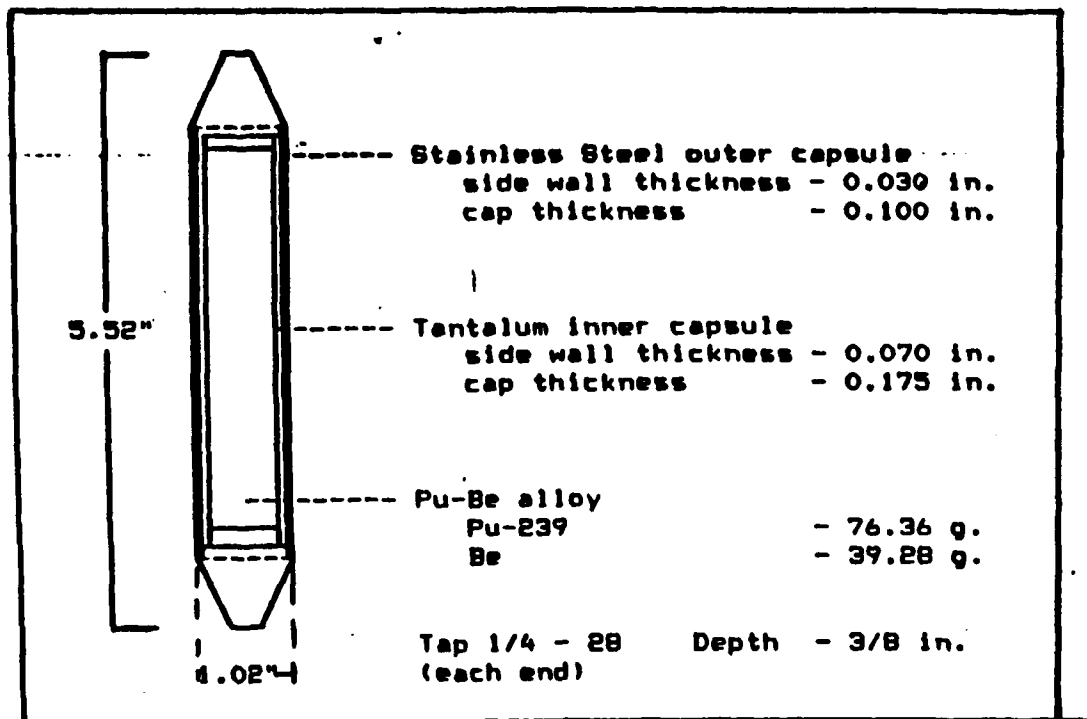


Figure 3-1. Dimensions of Pu-Be Source, M-1170 (13:8)

The term, "long counter" is given to this type of detector because it detects neutrons up to several MeV with a flat response, independent of neutron energy (17:321). Figure 3-2 shows the composition of the long counter built by Hanson and McKibben. The neutron source was first measured without the bath, to obtain a "bare source" value. Neutron leakage from the bath was then measured for each container size and concentration. The non-leakage factor was calculated by dividing the counts from the leakage measurement by the bare source counts and the source absorption term. The source absorption term was included to account for the extra fission neutrons caused by the bath. This measurement technique assumed that the source was isotropic. However, the source was not isotropic, which increased the uncertainty of the leakage calculations. The experimentally determined leakage was used only as a check for the leakage predicted from the computer model and was not used in the determination of Q .

Equipment

Table A-1, in the Appendix, lists the equipment used in the study.

Determination of Correction Factors

In this study, the computer model was used to calculate the source absorption, resonance absorption, and leakage terms. An input file was made to use in MORSE which modeled the source as a cylinder, surrounded by a cylinder of tantalum, and a cylinder of stainless steel. The endcaps of the stainless steel case were modeled as truncated right-hand cones. The manganese sulfate bath and polyethylene containers were modeled as cylinders. Using the *Combinatorial-Geometry* feature of the MORSE package, the components of the model were "assembled", and the source was located in the center of the manganese sulfate bath. The sizes used for the bath and the polyethylene containers were the same

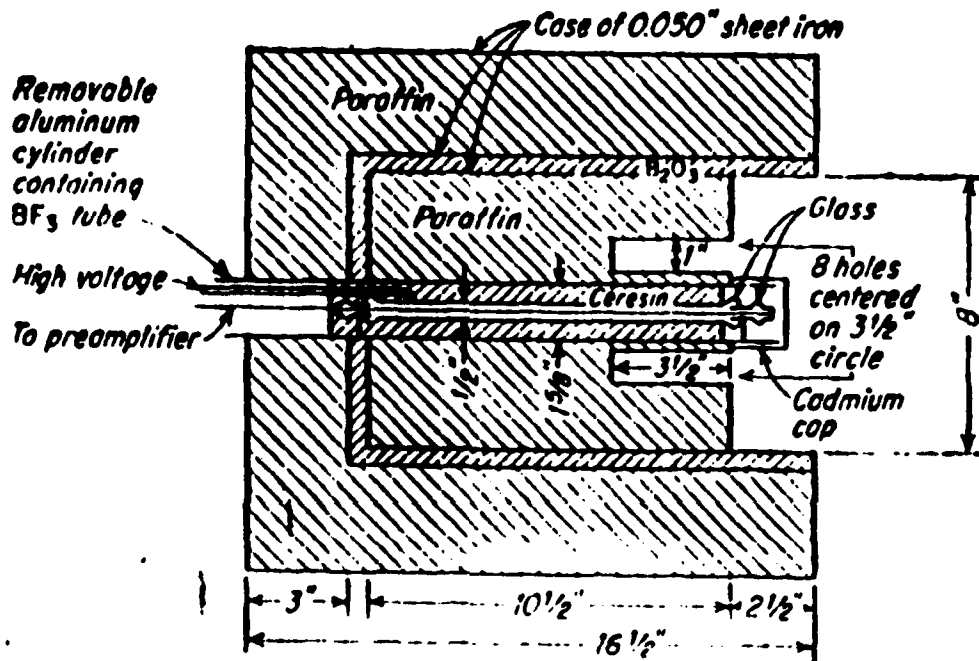


Figure 3-2. Hanson and McKibben "long Counter" (17: 321)

as the containers used in the experiment. A fourth container size was modeled to provide extra data between the 140 and 40 liter containers. Figure 3-3 shows the representation of the source generated by the program *PICTURE*, which was part of the MORSE package. Figure 3-4 shows the geometry of the source in the bath and polyethylene container, also generated by *PICTURE*. The cross-sections used for the MORSE calculations were obtained from DCL-31, a 37 neutron group cross-section tape, which was developed by Oak Ridge National Laboratory. These cross-sections, along with the number densities of the atoms in a particular medium, were in the program *XCHECKER* (a separate program that accompanied the cross-section library) to calculate cross-sections for that medium. The neutron source spectrum was chosen from Kumar and Nagarajan and adjusted to fit in the 37 group spectrum (11: 177). Figures 3-5 and 3-6 show the original and adjusted spectrum. The neutrons in each computer run were grouped in 50 "batches" of 200 neutrons each. The code package provided for the production of fission neutrons, and this option was used. The fission neutron spectrum was obtained from Bartine et al. (2: 11) Neutron histories were terminated by Russian roulette and escape.

A computer calculation was made with no bath present to determine how many fissions occurred normally in the source without the bath. The source absorption term was calculated for each of the manganese bath runs by dividing the total number of neutrons (source and fission neutrons) by the total number of neutrons from the bare source run. The leakage fraction was determined by dividing the weight of the neutrons that escaped by the total weight of the neutrons introduced. Since both the source and fission neutrons had an initial weight of 1, the total weight of the introduced neutrons was equal to the sum of the source neutrons and the fission neutrons. The manganese was found to have no significant absorption cross-section above the lower 5 energy groups of the

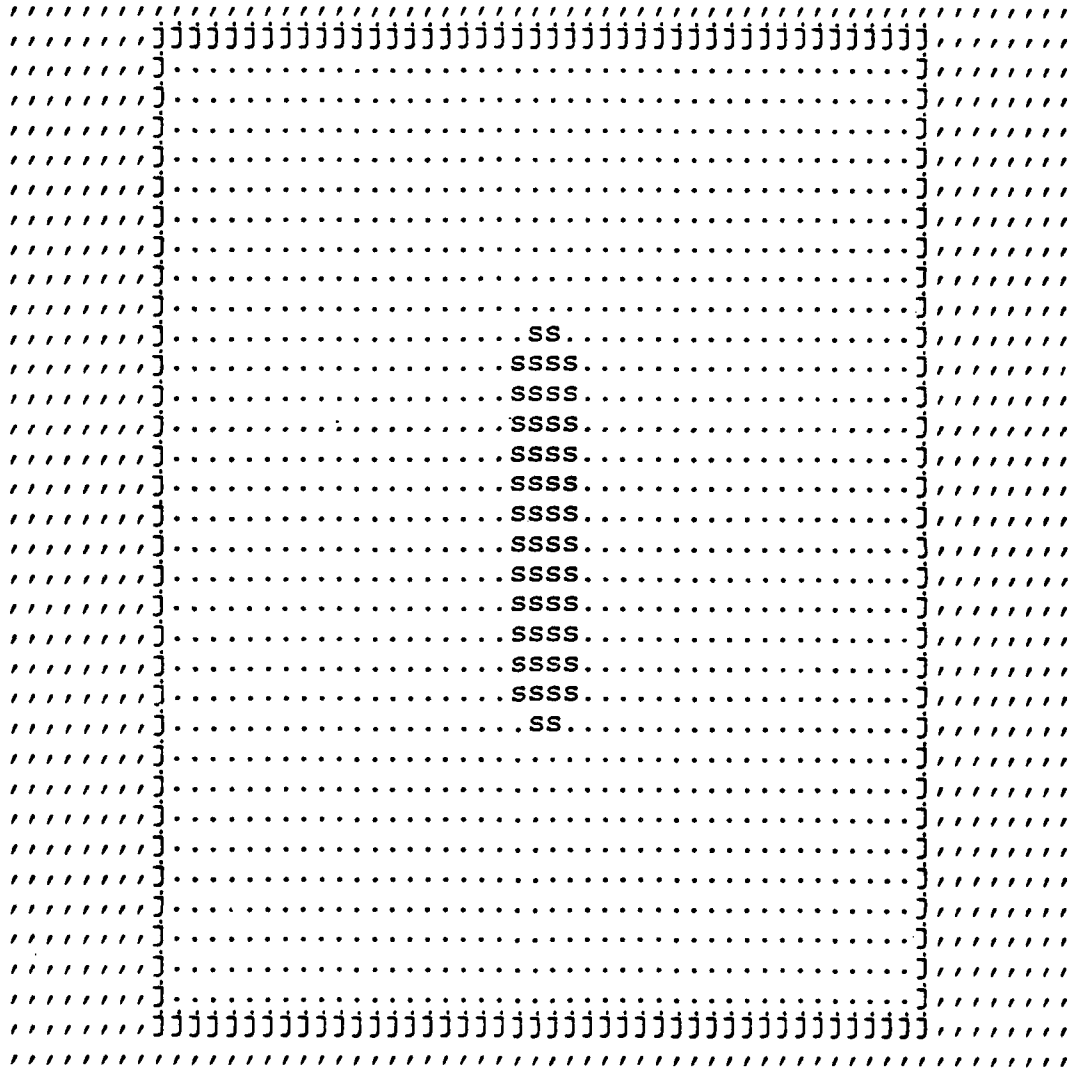


Figure 3-4. Geometry Generated by Picture

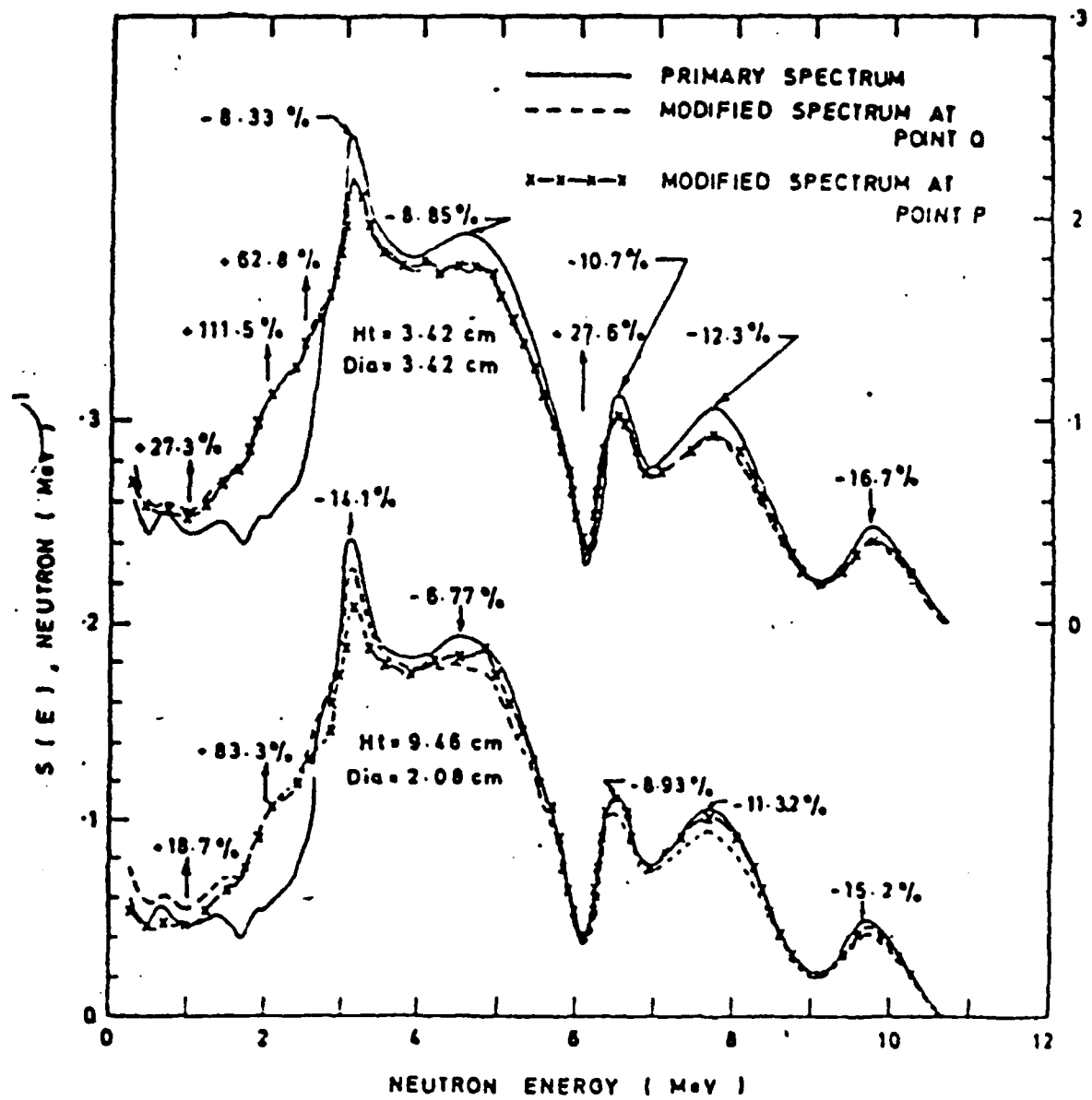


Figure 3-5. Pu-Be Neutron Source Spectrum (11:177)

Pu-Be Spectrum

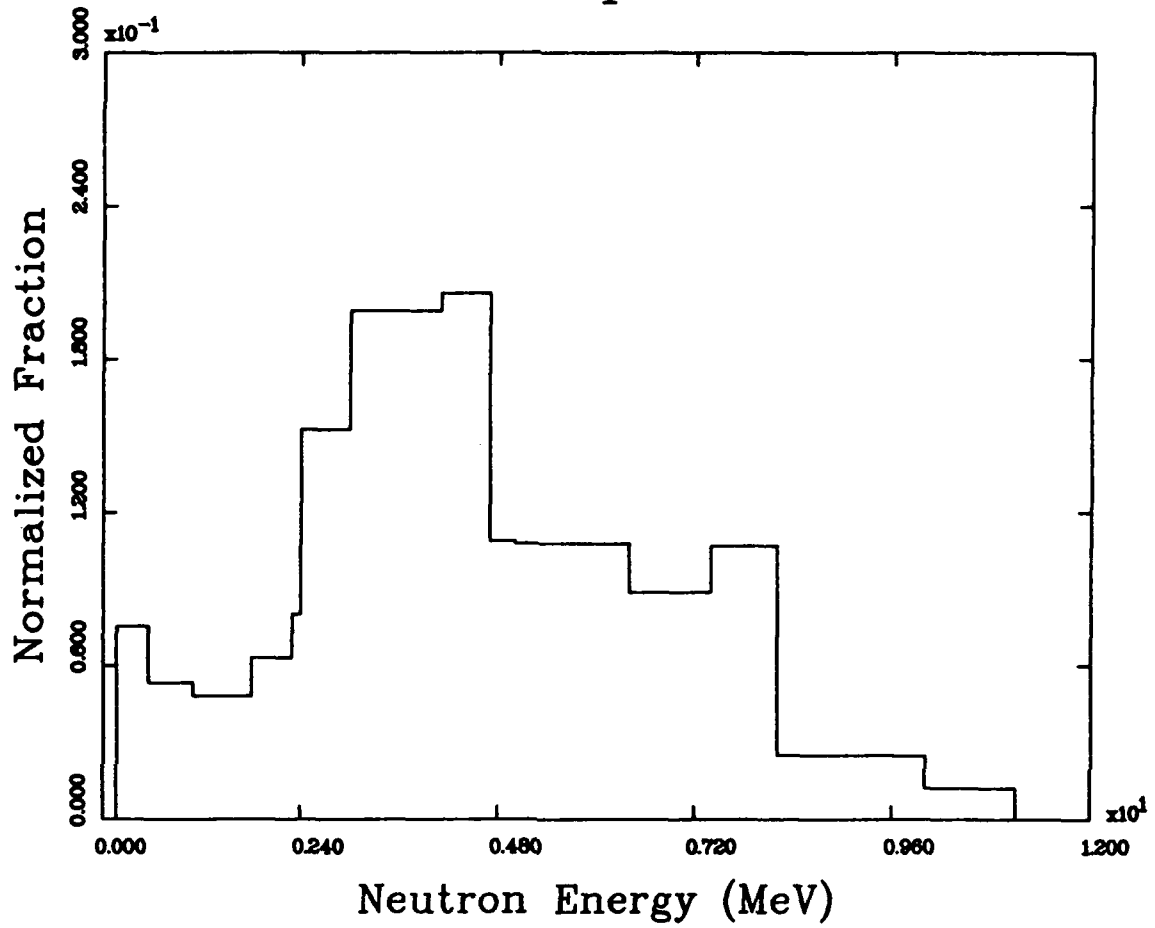


Figure 3-6. Source Spectrum Modified For MORSE

cross-sections (29 eV), so all of the absorptions in the higher 32 energy groups were considered to be absorbed by other nuclei. The resonance absorption fraction was calculated by dividing the total weight of all absorptions above 29 eV by the total weight of the source and fission neutrons.

The overall uncertainties in these factors were found by determining the random and systematic uncertainties involved in the calculations. The statistical uncertainties from the Monte Carlo calculations were considered random uncertainties. To account for the systematic uncertainties in the cross-section library, several uncertainties were included in the calculation of the overall uncertainties. The uncertainty in the ^{239}Pu thermal absorption cross-section was included in the source absorption term. The uncertainty of the leakage fraction included the 1.5% uncertainty in the thermal cross-section for ^{56}Mn . These estimates were considered to be conservative, but did not drastically increase the uncertainties. In the trials that included large container volumes, the uncertainties of the non-leakage factors were low because the leakage fraction was much lower than the non-leakage fraction. No adjustment was made to the uncertainty of the resonance absorption factor. Even a 10% uncertainty in the resonance absorption fraction would result in a negligible uncertainty in the resonance escape fraction when compared to the 1.5-1.9% uncertainties in several of the other terms, due to the 1.5% uncertainty in the ^{56}Mn absorption cross-section.

Determination of Solution Density

In order to obtain number densities of the nuclides in the bath, the density of the solutions had to be obtained. Metzler listed the volume of the MMC's used in his work (and this study) to be 5.685 +/- 0.005 liters (14: 44). The densities of the solutions were determined by dividing the masses of the solutions in the MMC's by this volume. In order to validate the densities obtained by these calculations,

a check was made of the 36.00% and the 5.56% solutions. One of the 17.8 liter containers was filled with distilled water to a point in the neck of the container. The volume of the container was calculated by dividing the mass of the water by the density of water at 20° C (18: F-11). The densities of the 36.00% and 5.56% solutions were determined by dividing the masses of the solutions, filled to the same point in the container, by the volume of the container. The densities of the 36.00% and 5.56% solutions determined from both methods agreed within 0.2%.

IV. RESULTS AND DISCUSSION OF RESULTS

In this section, results from the activation trials are presented, as well as the correction factors obtained from the computer model.

Results

Since several of the correction factors required for accurate source strength determination were difficult, if not impossible, to measure experimentally, no purely experimental estimates of source strength were made. The computer code was validated by checking several factors that could be measured more easily, or for which a value had been calculated by other researchers. The two major factors checked were neutron leakage and resonance absorption.

The comparison of the computer-modeled and experimentally measured leakage fractions is shown in Table 4-1. The agreement between computer predictions and experimental measurements differed by as much as 20%. However, the experimental measurements were made assuming that the same fraction of neutrons reached the BF_3 detector when the source was measured with the bath in place, as with the source measured with no bath. Since neither the source nor the bath were isotropic, the fractions of neutrons could differ significantly. Also, the measured leakage was calculated assuming a completely flat response for all neutron energies from the BF_3 detector. Price states that the "long counter has a flat response from 10 keV up to 3 MeV" (17: 321). The spectrum of the neutron source extends up to approximately 11 MeV. The efficiency of the counter may be different for neutron energies outside that range. A lower efficiency for the higher energy neutrons would account for the consistently higher values for the experimentally measured leakage. Therefore, the systematic uncertainty of the experimentally measured leakage was impossible to gauge, but could range as high as 5 to 10 percent. Only one of the six comparisons presented a significantly larger

Trial	Container Size (l)	MnSO ₄ Conc. (%)	Leakage Fraction	
			Computer Modeled	Exper. Measured
1	17.23	5.56	0.510 +/- 1.9%	0.551
2		9.00	0.494 +/- 1.9%	0.507
3	17.23	18.00	0.483 +/- 1.9%	0.505
4	17.23	36.00	0.444 +/- 1.9%	0.439
5	35.92	18.00	0.314 +/- 1.9%	0.332
6	35.92	36.00	0.289 +/- 1.9%	0.344

Table 4-1. Comparison of Leakage Fractions

difference between the measured and modeled leakage values.

The experimental measurement of resonance absorption was beyond the scope of this study. A value of 0.978 was estimated by Geiger and Whyte, based on measurements made by de Troyer and Tavernier (7: 260). All of the resonance absorption values generated by MORSE agreed with this estimate within 1 to 2 percent. Hwang and Choi obtained a correction for high energy, oxygen and sulfur capture of 2.88% + /- 0.5% for an Am-Be (α, n) neutron source (10: 71). This value would correspond to a 0.9712 + /- 0.015% resonance absorption term, which is also within 1 to 2 percent of the values obtained from MORSE.

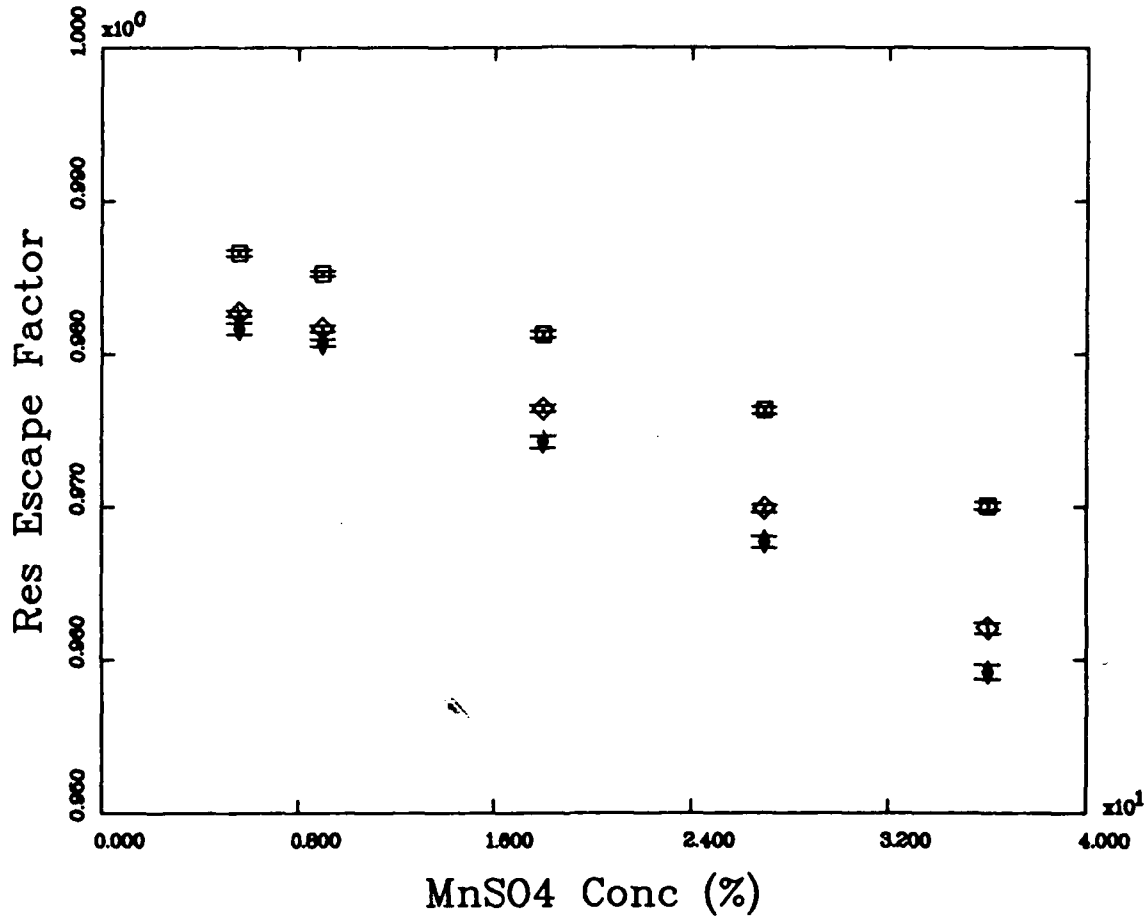
Table 4-2 is a list the resonance absorption correction factors used in this study. Figures 4-1 and 4-2 present how the resonance absorption factors differ with concentration (Fig. 4-1) and bath volume (Fig. 4-2). The MORSE generated values for high energy absorption indicate that the fraction of high energy absorptions increases as the $MnSO_4$ concentration increases, and increases slightly when the volume of the bath increases. This trend should be valid since increasing the $MnSO_4$ concentration increases the number density of sulfur and oxygen in the bath, and increasing the volume increases the path length that a high energy neutron must travel before escaping.

The agreement of the experimental values used as a check to the MORSE modeled values indicates that this computer model is valid for the geometries and other conditions used in this study. Table 4-3 is a list of the non-leakage factors calculated from the computer model. Figures 4-3 and 4-4 present the trends of the non-leakage factors as concentration and bath volume are varied. The higher concentrations of $MnSO_4$ tended to decrease neutron leakage since increasing the number density of ^{56}Mn increased the overall absorption cross-section of the bath. The extra absorbing medium from the larger bath volumes significantly reduced the fraction of neutrons that escaped.

Container Volume (liters)	Manganese Sulfate Concentration				
	36.00%	27.00%	18.00%	9.00%	5.56%
17.23	0.97006 +/-0.027%	0.97634 +/-0.024%	0.98129 +/-0.023%	0.98526 +/-0.017%	0.98664 +/-0.020%
35.92	0.96207 +/-0.043%	0.96989 +/-0.025%	0.97644 +/-0.022%	0.98166 +/-0.021%	0.98270 +/-0.019%
79.8	0.96172 +/-0.035%	0.96934 +/-0.032%	0.97570 +/-0.021%	0.98140 +/-0.028%	0.9833 +/-0.021%
158.0	0.95920 +/-0.047%	0.96769 +/-0.037%	0.97426 +/-0.039%	0.98074 +/-0.024%	0.98168 +/-0.038%
135.0			0.97120 +/-0.036%		

Table 4-2. Resonance Absorption Correction Factors

Resonance escape



□ --17.23 l
◇ --35.92 l
▼ --158.0 l

Figure 4-1. Resonance Absorption Factor

Resonance Escape

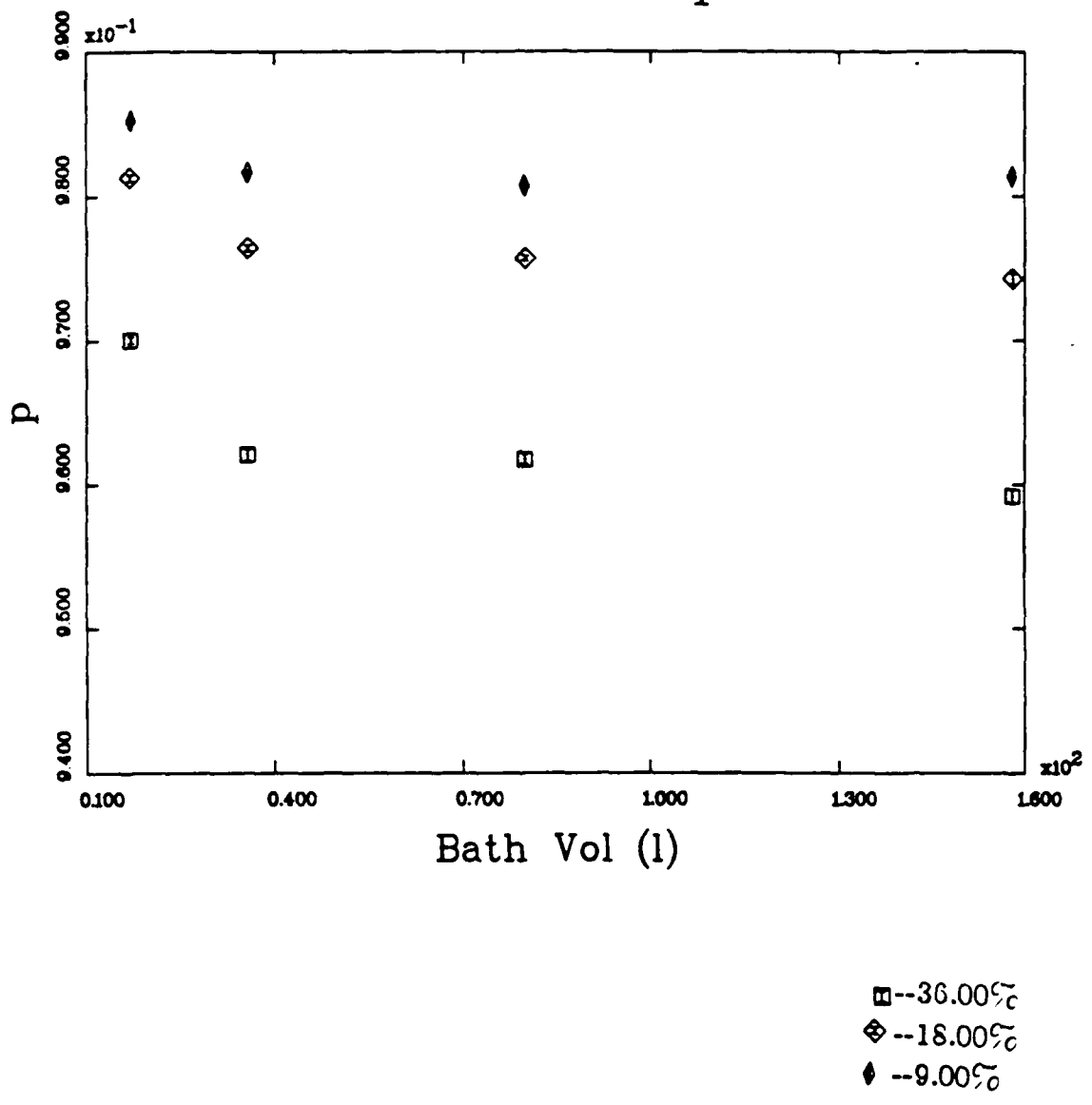
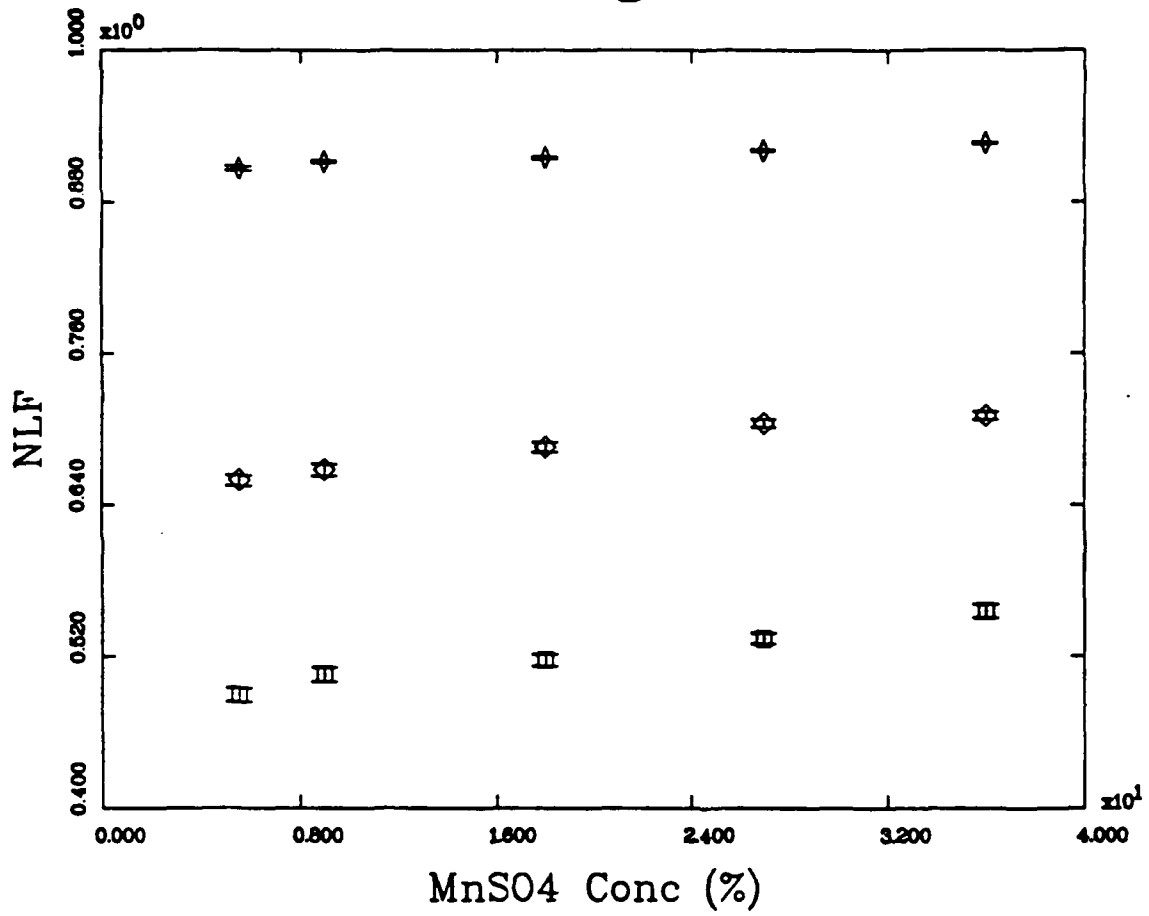


Figure 4-2. Resonance Absorption Factor

Container Volume	Manganese Sulfate Concentration				
	36.00%	27.00%	18.00%	9.00%	5.56%
17.23	0.558 +/-1.5%	0.534 +/-1.5%	0.517 +/-1.8%	0.506 +/-1.9%	0.490 +/-1.9%
35.92	0.709 +/-0.77%	0.705 +/-0.78%	0.686 +/-0.87%	0.668 +/-1.0%	0.659 +/-1.0%
79.8	0.857 +/-0.31%	0.848 +/-0.31%	0.837 +/-0.35%	0.831 +/-0.40%	0.818 +/-0.41%
158.	0.927 +/-0.14%	0.921 +/-0.15%	0.915 +/-0.18%	0.912 +/-0.18%	0.907 0.24%
135.			0.903 +/-0.17%		

Table 4-3. Non-Leakage Factors

Non-Leakage Factor

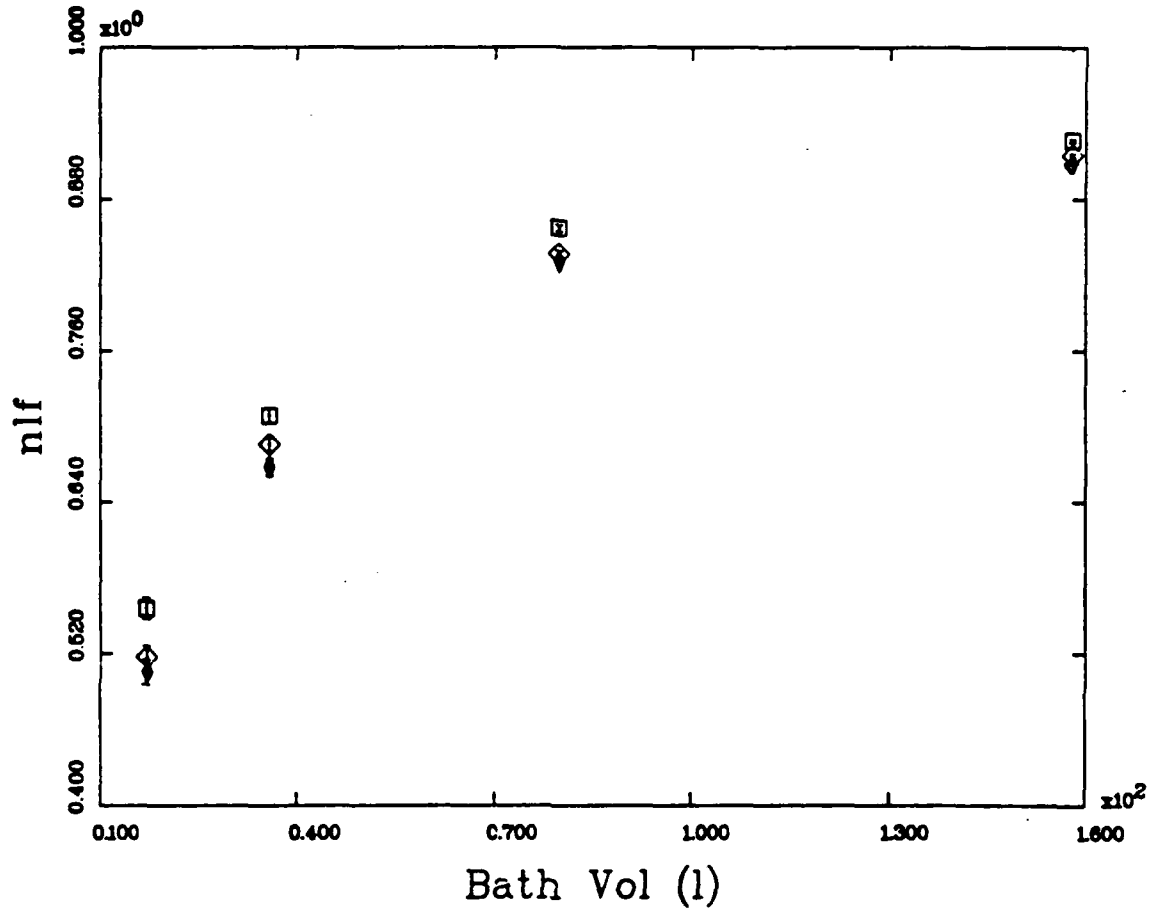


□ --17.23 l
◇ --35.92 l
△ --158.0 l

Figure 4-3. Non-Leakage Factor

Figure 4-3. Non-Leakage Factor

Non-Leakage Factor



□ --36.00%
◇ --18.00%
◆ --9.00%

Figure 4-4. Non-Leakage Factor

Table 4-4 is a list of the source absorption factors calculated in this study. Figures 4-5 and 4-6 show how the *sa* term varies with concentration and container volume. The fact that the *sa* remains almost constant with regards to bath volume indicates that the moderator is most important in the first few centimeters, since the source absorption term would go to one as the container volume went to zero. The extra absorbers in the higher concentrations of $MnSO_4$ tend to lower the *sa* term slightly, as shown by Fig. 4-6.

Figure 4-7 presents the values of the concentration adjustment factors for the detector efficiencies. The main sources of uncertainty are the random uncertainty in the counting data and the systematic uncertainty from weighing the masses of the solutions and measuring the activities.

Figure 4-8 presents the values for the *MAF*. The uncertainties in these values are primarily from the uncertainties in the cross-sections of ^{55}Mn (1.6%) and sulfur (5.8%). The uncertainties in the relative number densities of the atoms were less than 0.3%, and did not significantly increase the overall uncertainties.

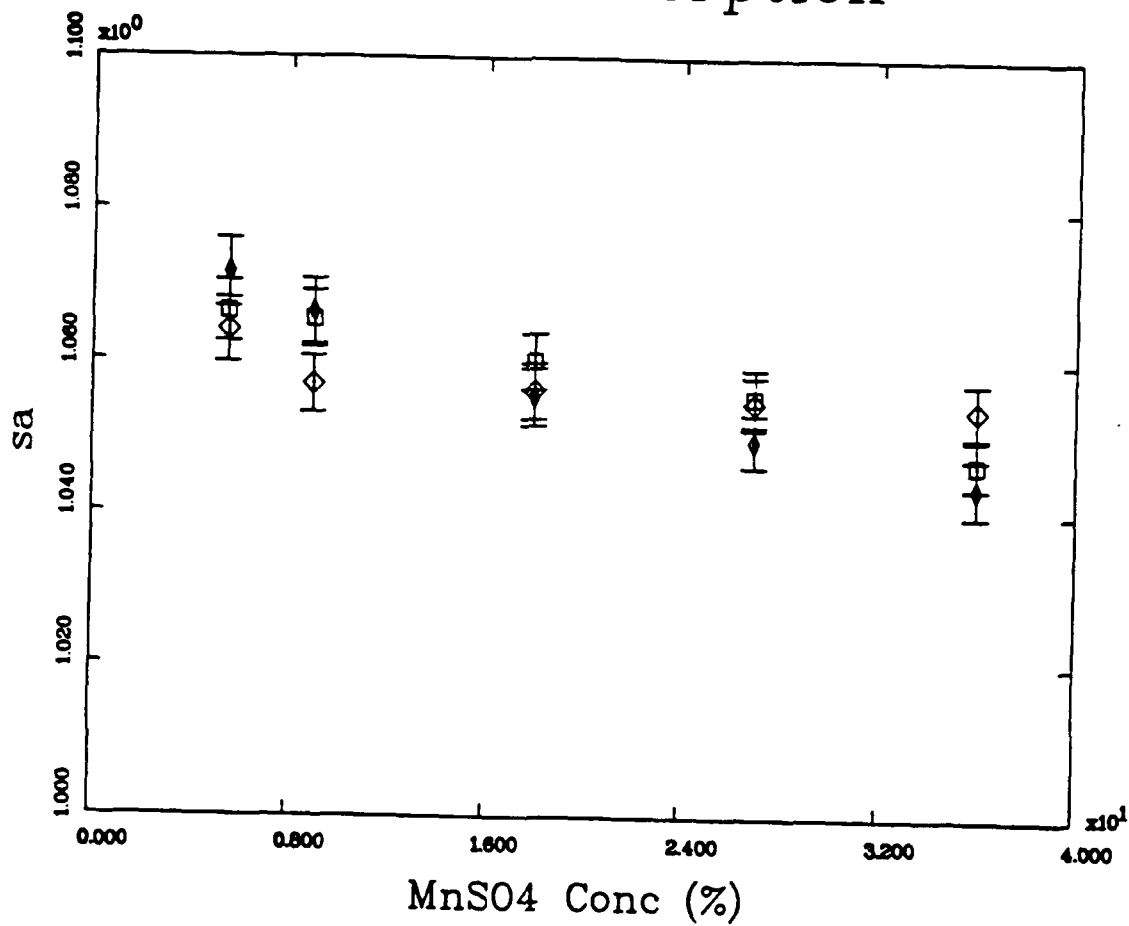
Table 4-5 presents the total neutron output of M-1170 determined from the activation trials and computer generated correction factors. One trial was rejected (trial #9) when calculating the statistical average of the neutron source strength. The total neutron output of the neutron source was determined to be 1.072×10^7 +/- 3.1% neutrons per second. The uncertainty decreases slightly as the bath volume increases due to the lower uncertainty of the non-leakage factor. The neutron output calculated from the from the product specifications that accompanied the source was 1.094×10^7 neutrons per second which is within the uncertainty of the experimental prediction.

While random uncertainty occurred in the activity measurements, the concentration adjustment factor, and the Monte Carlo calculations, the largest part of the uncertainties encountered in this study were systematic.

Container Volume (liters)	Manganese Sulfate Concentration				
	36.00%	27.00%	18.00%	9.00%	5.56%
17.23	1.047 +/-0.51%	1.055 +/-0.54%	1.060 +/-0.53%	1.066 +/-0.53%	1.067 0.56%
35.92	1.047 +/-0.52%	1.055 +/-0.52%	1.058 +/-0.53%	1.057 +/-0.53%	1.064 +/-0.57%
79.8	1.051 +/-0.55%	1.054 +/-0.52%	1.058 +/-0.56%	1.067 +/-0.57%	1.060 +/-0.57%
158.0	1.044 +/-0.54%	1.050 +/-0.52%	1.057 +/-0.58%	1.067 +/-0.57%	1.072 +/-0.58%
135.0			1.055 +/-0.54%		

Table 4-4. Source Absorption Factors

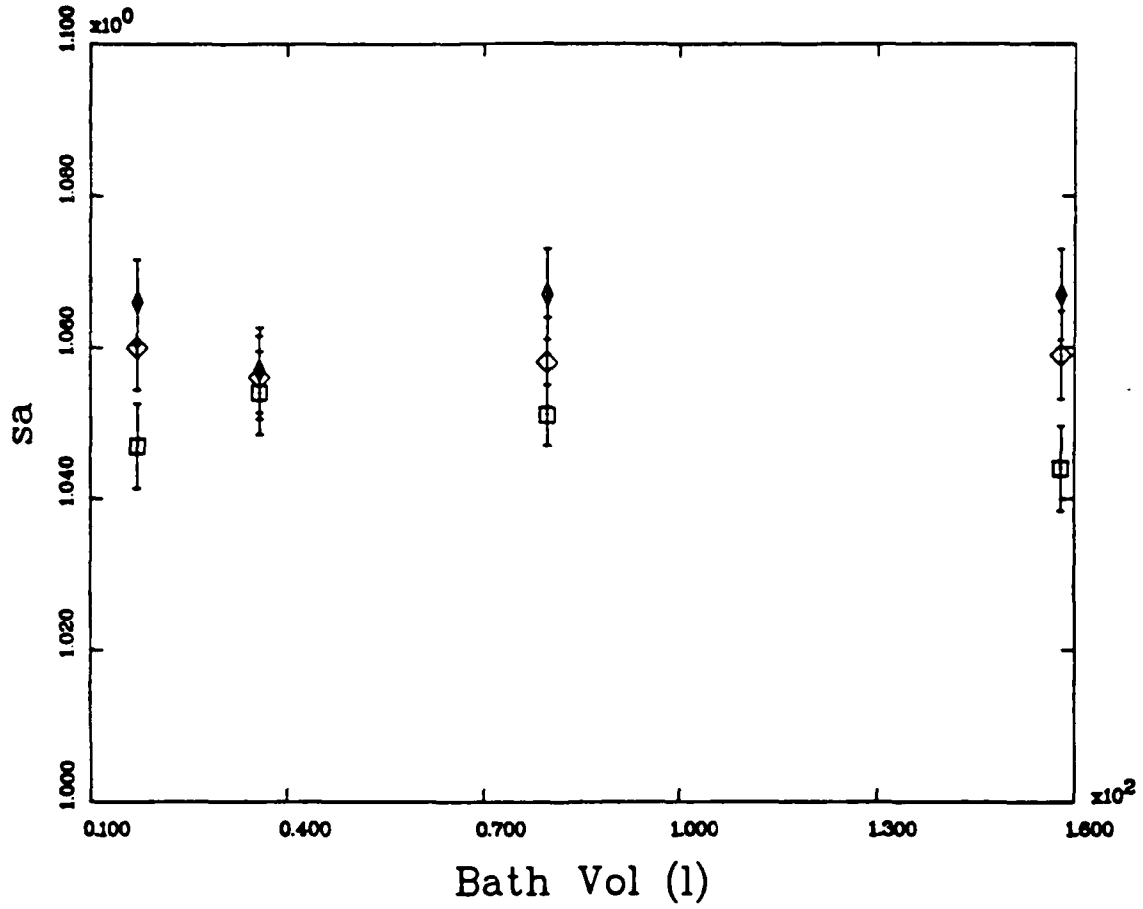
Source Absorption



□ --17.23 l
◇ --35.92 l
◆ --158.0 l

Figure 4-5. Source Absorption Factor

Source Absorption



□ --36.00%
◇ --18.00%
◆ --9.00%

Figure 4-6. Source Absorption Factor

Conc Adj Factor

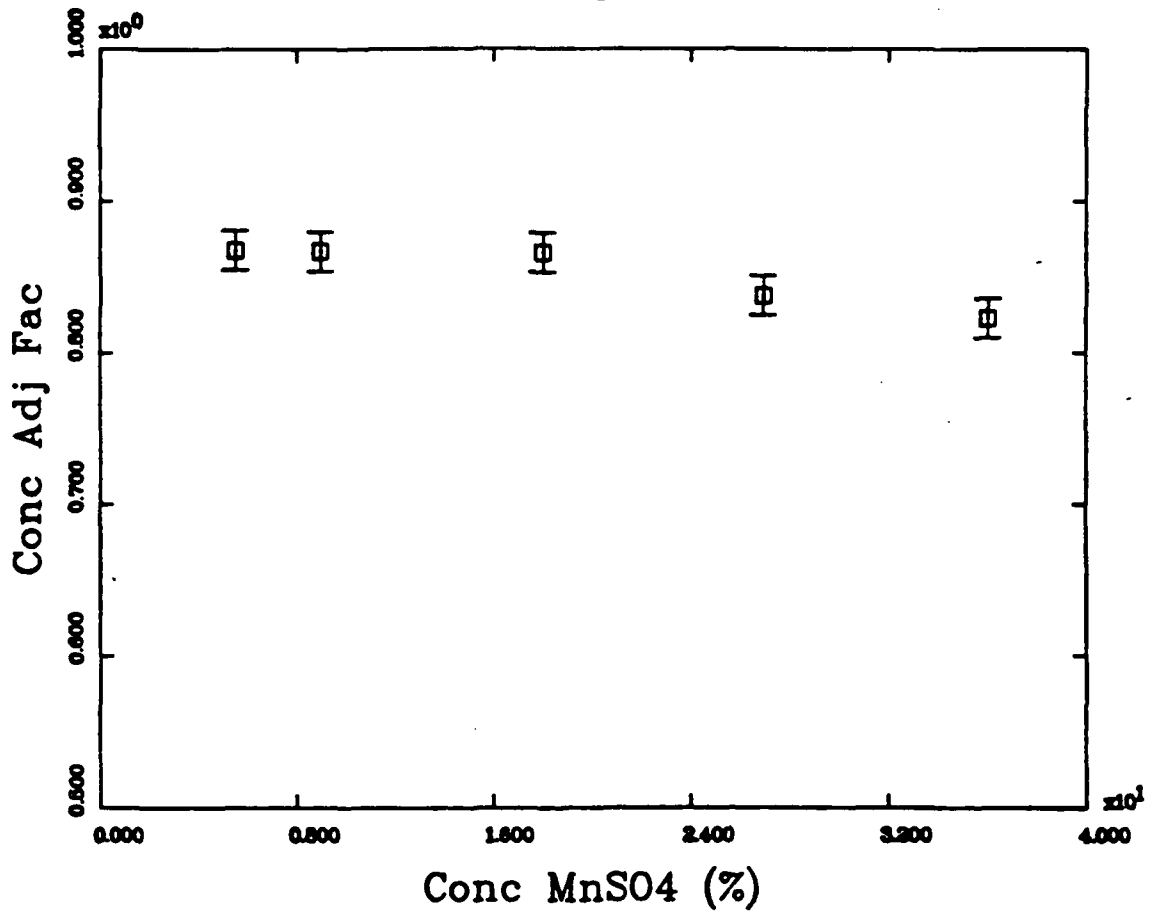


Figure 4-7. Concentration Adjustment Factors

MAF

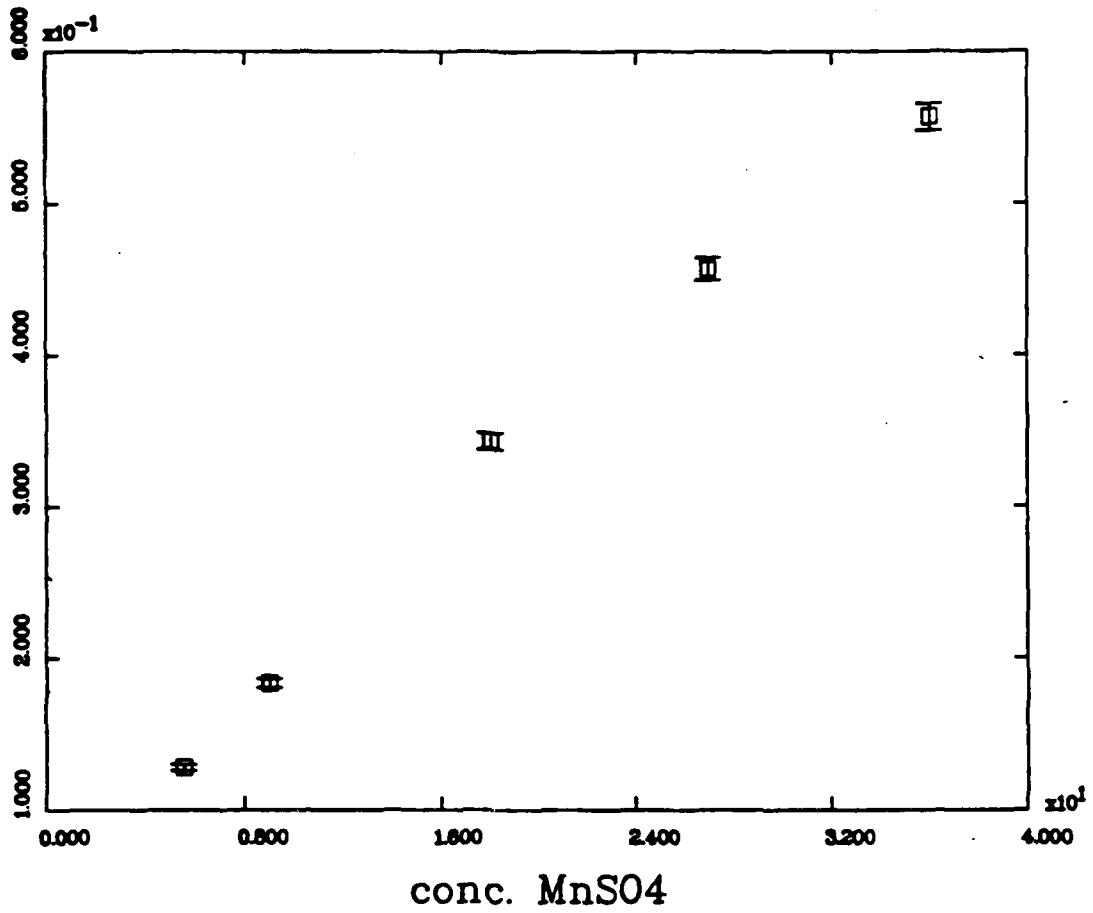


Figure 4-8. Manganese Absorption Fraction

Trial	Bath	Source Strength	
1	17.23 ± 5.56%	1.079×10^7	+/- 3.1%
2		1.085×10^7	+/- 3.1%
3	17.23 ± 9.0%	1.089×10^7	+/- 3.1%
4		1.067×10^7	+/- 3.1%
5	17.23 ± 18.0%	1.071×10^7	+/- 3.1%
6		1.051×10^7	+/- 3.1%
7	17.23 ± 36.0%	1.018×10^7	+/- 3.1%
8	35.92 ± 36.0%	1.062×10^7	+/- 2.6%
9	35.92 ± 18.0%	9.30×10^6 *	+/- 2.6%
10		1.073×10^7	+/- 2.6%
11	135 ± 18.0%	1.136×10^7	+/- 2.5%

Table 4-5. Neutron Source Strength

The largest source of systematic uncertainty was the 1.6% uncertainty in the *MAF* caused by the uncertainty in the thermal cross-sections of manganese and sulfur. Mughabhab and Garber list thermal cross-sections of 13.3 +/- 0.2 and 0.52 +/- 0.030 barns for manganese and sulfur, respectively. Arbildo presented values of 13.4 +/- 0.1 and 0.531 +/- 0.015 barns for thermal cross-sections of manganese and sulfur (16: 80 + 87). Had these values been used in this study, the systematic uncertainty in the *MAF* could have been reduced to 1.1%, and overall uncertainty in each trial could have been reduced by approximately 0.3%. The older values were used in this study, mainly because the cross-section tape used for the computer model predated Arbildo's work and used the older values.

V. CONCLUSIONS AND RECOMMENDATIONS

Activation of manganese sulfate is a simple yet precise method of determining the source strength of a neutron source. The initial goal of this study was to reduce the overall uncertainty in the method to less than 3%, and this has been done. The precision of the overall estimate of the activity could be improved by refining the concentration adjustment factor. An accurate efficiency could be obtained by preparing the liquid radionuclide standard in a given concentration of manganese sulfate. However, while the calibration should be very accurate at that concentration, the efficiency of the detection system would be valid only for that concentration. This study sought to provide a calibration method that would be valid for a wide range of manganese sulfate baths, and the inclusion of the concentration adjustment factor was the easiest method to accomplish that goal. More precise (smaller uncertainty) cross-sections, such as those presented by Arbildo, in the computer model as well as the MAF, could also improve the precision of this method.

The computer model of this experiment could be extended to other types of neutron sources with little modification. The geometry of the source could be modeled by changing the geometry in the *Combinatorial Geometry* portion of the input file used for the computer code. The source spectrum would also have to be known, as well as the fission spectrum, if any. These spectra would have to be fit to the energy group divisions of the cross-section file. The subroutine *SOURCE* would also have to be modified to match the distribution of neutron production in the source. If the source produced fission neutrons, as does M-1170, a "bare source" calculation would be required to determine what fraction of the normal source spectrum is due to fission. If the source did not produce fission neutrons, then a detector would be needed to calculate absorptions in the source. This could be done by modifying detector 1 in the code to calculate absorptions, similar

to the manner detector 2 (manganese bath absorption) was modified in this study. A cross-section file, or tape which contained all of the nuclei involved in the model would also be required. The tape used in this study, DCL-31, contained 37 neutron groups and fission cross-sections for the Plutonium. A tape containing 20 to 50 neutron groups would probably produce sufficient results. Further modifications would depend on the requirements and imagination of the individual researcher.

Appendix A: Equipment List

Major Items of Equipment			
Item	Manufacturer	Model	Serial #
Detector Ge(li)	Princeton Gamma Tech	LGC17HT	1609
HV Pwr Sup	Nim Std	AEC-5000	PL 751
Preamp	Princeton Gamma Tech	RG-11AC	2982
Lin Amp	ORTEC	452	
MCA	Nuclear Data	ND-66	
		ND-680	96-1447
Neutron Dosimeter	Texas Nuclear	9146	PL 607
Long Counter	AFIT		
Preamp	ORTEC	109 PC	
Lin Amp	ORTEC	440A	PL 768
SCA	ORTEC	406A	1022
Timer/Scaler	Tennelec	562P	1871191
Oscilloscope	Tektronix	465M	B054149
Neutron Source	Mound Lab	Pu-Be	M1170
Furnace	Thermalyne	1500	F-C1525M
Precision balance	Right-A-Weigh		37785
Beam Balance	Ohaus		1201
Bench Balance	Horns		3108
Computer	Digital Equip.	Vax	
Computer	RSIC	MORSE	
Code			

Table A-1

Appendix B: Number Densities Used In Computer Model

ELEMENT	NUMBER DENSITY (atoms/barn · cm)
Source	
⁹ Be	8.501 x 10⁻²
²³⁹ Pu	6.326 x 10 ⁻³
¹⁸¹ Ta	5.527 x 10 ⁻²
Cromium	1.743 x 10 ⁻²
Manganese	1.740 x 10 ⁻³
Iron	5.936 x 10 ⁻²
Nickel	7.720 x 10 ⁻³

Manganese Sulfate Solution

	36.00%
Hydrogen	6.323 x 10 ⁻²
Manganese	2.099 x 10 ⁻³
Oxygen	4.001 x 10 ⁻²
Sulfur	2.099 x 10 ⁻³
	27.00%
Hydrogen	6.495 x 10 ⁻²
Manganese	1.418 x 10 ⁻³
Oxygen	3.815 x 10 ⁻²
Sulfur	1.418 x 10 ⁻³
	18.00%
Hydrogen	6.654 x 10 ⁻²
Manganese	8.778 x 10 ⁻⁴
Oxygen	3.833 x 10 ⁻²
Sulfur	8.878 x 10 ⁻⁴

Manganese Sulfate Solution (Cont'd)

9.00%

Hydrogen	6.779×10^{-2}
Manganese	3.779×10^{-4}
Oxygen	3.491×10^{-2}
Sulfur	3.779×10^{-4}

5.56%

Hydrogen	6.689×10^{-2}
Manganese	2.478×10^{-4}
Oxygen	3.443×10^{-2}
Sulfur	2.478×10^{-4}

Polyethylene Container

Hydrogen	7.900×10^{-2}
Carbon	3.950×10^{-2}

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Vita

Paul Fletcher Whitworth Jr. was born on November 4, 1959, in Rome Georgia, and is the son of Fletcher and Elnora Whitworth. He graduated from North Cobb High School in Acworth Georgia in June 1977. He received his Bachelor of Science degree from Mercer University in Macon, Georgia, where he majored in Physics and Biology, minoring in Chemistry. He received a commission in the United States Army Chemical Corps in June 1981. He was assigned to Fort Carson, Colorado, where he served as a Battalion Chemical Officer and a Chemical Platoon Leader in the Fourth Infantry Division, and as Group Chemical Officer and Headquarters Company Commander with the 43d Support Group. While assigned to the 43d, he participated in Operation Ahuas Tara II in Honduras. After attending the Chemical Officer Advanced Course in Fort McClellan, Alabama, he entered the Air Force Institute of Technology. He and his wife Ruth have one son, Stephen.

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ABSTRACT

In this study the manganese-sulfate-bath technique was used to determine the total neutron output of a Plutonium-Beryllium neutron source. Activation trials were conducted using 4 different concentrations of manganese sulfate and 3 different size containers. Correction factors for source absorption, non-leakage, and resonance absorption were calculated using data generated by *MORSE*, a Monte Carlo neutron transport code. Values for these factors were calculated for manganese sulfate concentrations between 5.56% and 36.00%, and volumes from 17.2 to 160 liters in cylindrical containers. A source strength of $1.07 \times 10^7 \pm 3\%$ neutrons per second was found by using experimentally measured manganese activity in conjunction with the computer-predicted correction factors.

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