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**THE INEL BERYLLIUM MULTIPLICATION EXPERIMENT
FINAL REPORT**

J. R. Smith
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THE INEL BERYLLIUM MULTIPLICATION EXPERIMENT
FINAL REPORT

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ABSTRACT

The experiment to measure the multiplication of 14-MeV neutrons in bulk beryllium has been completed. The experiment consists of determining the ratio of ^{56}Mn activities induced in a large manganese bath by a central 14-MeV neutron source, with and without a beryllium sample surrounding the source. The superior isotropy and flat energy response of the manganese bath gives this detector an advantage over the inhomogeneous and anisotropic detector arrays used in previous experiments for measurements of this type. Values of the multiplication have been obtained for beryllium samples of four thicknesses. For beryllium thicknesses of 4.6, 12.1, 15.6, and 19.9 cm the apparent multiplication values are 1.390, 1.928, 2.072, and 2.127, respectively. These values are affected by several systematic effects characteristic of the manganese bath. The values of these systematic corrections are established by a combination of calculation and experimental parameterization. The corrected net multiplication values for pure beryllium are 1.311, 1.764, 1.904, and 1.988, respectively. Detailed calculations of the multiplication and all the systematic effects were made with the Monte Carlo program MCNP, utilizing both the Young and Stewart and the ENDF/B-VI evaluations for beryllium. Both data sets produce multiplication values that are in excellent agreement with the measurements for both raw and corrected values of the multiplication. We conclude that there is no real discrepancy between experimental and calculated values for the multiplication of neutrons in bulk beryllium.

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INTRODUCTION

Beryllium is being considered for use as a neutron multiplier to improve the neutron economy and tritium breeding ratio of several types of fusion systems. Because beryllium has a relatively high (n,2n) cross section with a low threshold, it should be the most effective multiplier that can be used. However, integral experiments by Cloth¹, Basu et al.^{2,3}, and Hartley et al.^{4,5} indicate that the effective multiplication of 14-MeV neutrons in bulk beryllium may be lower, by perhaps 20%, than that calculated from microscopic data. On the other hand, the data of Wong et al.⁶ and an old manganese bath measurement at Lawrence Livermore National Laboratory⁷ both indicate multiplication values as large as, or greater than, those predicted by microscopic data. The uncertainty indicated by the range of experimental values is of critical importance to the utility of beryllium as a neutron multiplier. None of these previous experiments has attained the accuracy required to resolve the discrepancy. However, Chen⁸ has recently reported a new measurement of the neutron multiplication in beryllium. He claims an accuracy of 2.8%, and observes multiplication values 15% lower than those calculated using ENDF/B-VI data. It seemed appropriate to perform a new measurement using the manganese bath, employing techniques that have been refined in many respects since the Livermore experiment⁷ was performed.

EXPERIMENTAL ARRANGEMENT

Principle of the Measurement

The manganese bath is the preferred method for the measurement of the absolute value of neutron source strengths. It has a history of success in the measurement of neutron multiplicities for ²⁵²Cf and the fissile nuclei⁹, as well as for the measurement of emission rates of neutron sources of various energy spectra.¹⁰ It has the advantages that it is a 4 π detector, and its response is very uniform as a function of both angle and energy of the neutrons. The effect of these advantages is that the manganese bath may detect neutrons that may be missed by other detection methods.

In the manganese bath method a neutron source is placed at the center of a totally-absorbing aqueous solution of MnSO_4 . The capture of neutrons by Mn produces a ^{56}Mn activity proportional to the emission rate of the source. As applied to the measurement of the multiplication of 14-MeV neutrons in bulk beryllium, the neutron source is a tritium target placed at the end of the drift tube of a small deuteron accelerator. Surrounding the source is a sample chamber. When the sample chamber is empty, the neutrons go directly to the surrounding MnSO_4 solution, and produce a ^{56}Mn activity proportional to the neutron emission rate. When the chamber contains a beryllium sample, the neutrons first enter the beryllium and multiply through the (n,2n) process. Neutrons escaping from the beryllium enter the bath and produce a ^{56}Mn activity proportional to the neutron emission rate multiplied by the effective value of the multiplication in bulk beryllium. The ratio of the activities with and without the sample present is proportional to the multiplication value. Irradiations are monitored by and normalized to the counting rate of the associated α -particle detector.

Manganese Bath

The manganese bath assembled for the beryllium multiplication experiment is shown in Figure 1. It utilizes a polyethylene tank, basically cylindrical in shape, but with an ellipsoidal domed top. The inside diameter of the tank is approximately 181.9 cm and the inside maximum height is 188.9 cm. The wall thickness is 1.27 cm. Shown with the tank is the sample chamber, which is an aluminum cylinder of inside diameter and height 60.96 cm, mounted on aluminum legs and a supporting base of stainless steel. The structure is assembled inside the tank.

The MnSO_4 solution is circulated by two Crane Series 800 Dynapumps. One of these is the mixing pump. It continuously pumps solution from the lower part of the bath to a pair of diffusing nozzles near the top of the bath, at a rate of about 2.5 liters per second. The second pump is the flow pump. It continuously draws solution from the lower part of the bath, at a position opposite that of the mixing pump, and delivers it to the detectors, then returns it to the top of the bath. The flow rate to each detector is about 0.27 liters

Figure 1. Manganese Bath and Sample Chamber

per second. The detectors are a pair of 7.6 cm x 7.6 cm NaI(Tl) scintillation counters. Counting is done continuously, during both the growth and decay phases of the irradiation.

Neutron Generator

Figure 2 shows the neutron generator installed at the manganese bath. The banding now seen on the tank minimizes bulging as the tank is filled, and preserves the alignment of the neutron generator. The generator is mounted upon a trolley that allows the neutron source to be placed at the center of the bath for irradiations or withdrawn for change of samples. The drift tube and target chamber are based upon the low-mass design developed at Livermore for their pulsed-sphere measurements¹¹. An associated-particle detector is mounted at 174° to the incident beam. It is located in the underslung portion of the cone seen at the end of the drift tube in Figure 2.

Figure 2. Manganese Bath and Neutron Generator

The most stable mode of operation of the neutron generator is to maintain the machine potential at a constant 150 kV. The machine has a Penning ion source and no beam analyzer. Therefore the beam arriving on target is a mixture of atomic and molecular ions. A typical spectrum of associated alpha particles is shown in Figure 3. The principal peak represents alpha particles from (d,t) collisions involving the molecular beam. Analysis of the pulse-height spectrum from the α -detector indicated that approximately 97% of the (d,t) ions in the beam were molecular, with the remainder being atomic. The deuteron beam energy is therefore 97% 75 keV and 3% 150 keV. Typically the generator produces approximately 10^8 14-MeV neutrons per second.

A single-channel analyzer accepts pulses under the alpha peak for monitoring of the neutron yield of the source. The background under the alpha peak is approximately 0.75% of the foreground. Since the data are ultimately used in the form of ratios of activities, the small background is of negligible consequence.

Figure 3. Typical α -particle spectrum from associated-particle detector.

Beryllium Samples

The beryllium samples used in this experiment were obtained on loan from Lawrence Livermore Laboratory (LLL). Table 1 describes the physical dimensions of the samples. They are the same set of beryllium spherical shells used in the Hartley^{4,5}, and Wong⁶ measurements, with one exception. The experiments of Hartley et al. and Wong et al. were handicapped by having a void in the position represented by the shell designated L-8 in Table 1. The original beryllium shells fabricated for this part of the set have been missing for many years. Through the efforts of Ralph Moir of LLL and Karl Porges of ANL a replacement shell for this position has finally been fabricated. A set of beryllium blocks, 15.2 cm x 15.2 cm x 7.6 cm, was provided by ANL. The LLL shop cemented these blocks together, using epoxy cement, to form a beryllium monolith from which piece L-8 was machined. This piece contains a quantity of epoxy cement

TABLE 1. LLL BERYLLIUM HEMISPHERICAL SHELLS

Sample	Shell No.	A. Left Hemisphere		Thickness (cm)	Mass (grams)
		IR (cm)	OR (cm)		
Be #1	L 1	7.992	8.630	0.638	475.4
	L 2	8.530	9.140	0.511	437.3
	L 3	9.136	9.779	0.643	626.9
	L 4	9.779	10.287	0.508	558.1
	L 5	10.286	10.921	0.635	790.2
	L 6	10.922	11.557	0.635	896.9
	L 7	11.558	12.572	1.013	1658.0
Be #2	L 8	12.576	20.077	7.501	22955.0 ^a
Be #3	L 9	20.084	21.844	1.760	8877.0
	L 10	21.844	23.622	1.778	10399.0
Be #4	L 11	23.618	25.396	1.778	12233.0
	L 12	25.399	27.934	2.535	20732.0
Totals Left Hemisphere				19.935	80638.8
Sample	Shell No.	B. Right Hemisphere		Thickness (cm)	Mass (grams)
		IR (cm)	OR (cm)		
Be #1	R 1	8.002	8.310	0.307	239.4
	R 2	8.316	8.631	0.315	260.7
	R 3	8.637	9.140	0.503	461.9
	R 4	9.145	9.775	0.630	658.7
	R 5	9.782	10.284	0.503	585.7
	R 6	10.293	10.921	0.627	822.8
	R 7	10.927	11.557	0.630	931.6
Be #2	R 8	11.561	12.572	1.011	1719.9
Be #3	R 9	12.579	15.239	2.659	6000.0
	R 10	15.245	20.066	4.821	17594.0
Be #4	R 11	20.079	21.844	1.765	8934.0
	R 12	21.857	23.619	1.763	10670.0
Be #4	R 13	23.631	25.391	1.760	12311.0
	R 14	25.411	27.936	2.525	21019.0
Totals Right Hemisphere				19.819	82208.7
Total Sample Mass					162847.5

^a Shell L 8 mass includes approximately 84.72 g D-85 cement.

calculated to be 84.72 grams. The epoxy is included in the list of impurities in the sample. The presence of the L-8 shell greatly improves the experiment. Without it we would have been required either to leave out the shells on the right-hand side opposite the void, or to fill the void with an aluminum can containing BeO. Either of these steps would have posed severe problems in the performance and interpretation of the experiment.

Table 1 shows that there are a total of 12 shells in the hemisphere through which the drift tube of the neutron generator enters, and 14 shells in the opposite hemisphere. The horizontal lines demark the grouping into the four experimental samples used. The left-hand shells are lighter than the right-hand shells because of they contain the entrance holes for the drift tube of the neutron generator. The assembled dimensions of the four experimental samples are described in Table 2. In keeping with the variations in dimension among the

TABLE 2. BERYLLIUM EXPERIMENTAL SAMPLES

Sample No.	IR (cm)	OR (cm)	Thickness (cm)	Mass (kg)	Fraction Solid
Be #1	8.0	12.6	4.6	11.12	0.9791
Be #2	8.0	20.1	12.1	57.67	0.9850
Be #3	8.0	23.6	15.6	96.55	0.9880
Be #4	8.0	27.9	19.9	162.85	0.9905

shells the dimensions are rounded off to one decimal place and the mass to two decimal places. Table 2 also lists the "Fraction Solid" for the four samples. This is the ratio of the mass of the actual sample to the mass that would be present without the entrance hole for the drift tube. The values were calculated by assuming that the left-hand shells would have the same mass as the right-hand shells if the holes were not present. The calculation neglects the effect of several small holes cut in the samples for alignment pins and foil irradiation positions. The four assembled experimental samples are pictured in Figure 4. Each sample sits on its own stand, which is of aluminum with holes punched to

Figure 4. The four beryllium samples. From left to right: Be #1, Be #2, Be #3, and Be #4.

decrease the neutron absorption of the structure.

Because the LLL samples were fabricated many years ago, no record of a chemical analysis of these specific pieces could be located. When piece L-8 was fabricated, however, an analysis was made of one of the scrap pieces. The results of this analysis appear in Table 3. We assume that the composition of the older samples was similar to that of L-8. Therefore the Table 3 data were used for all of the beryllium samples used in this experiment.

The mechanical problems associated with placing a 162.8-kg beryllium sample in proper alignment at the center of the manganese bath and removing it proved challenging. It is important to keep the structural materials as light as possible to minimize neutron absorption, while providing adequate support for the

heavy samples. The largest beryllium sample consists of twenty-six hemispherical shells. Entrance holes for the neutron generator drift tube are at an angle of 45° to the equator for the inner shells, and 90° for the outer shells. The problem is to assemble these shells into an array whose final orientation has the equator of the outer shells 3° off of the vertical, with the equator of the inner shells at 45° to the rest of the sample. The 3° offset is to allow for the underslung position of the α -particle detector. After assembly the whole sample must be lifted above the manganese bath and lowered into the sample chamber in such a manner that the holes are properly aligned to allow entrance of the drift tube of the neutron generator. The mechanism devised for handling these tasks is illustrated in Figures 5 through 7. The apparatus was adapted from a tilting cradle for 208-liter drums. An alignment spindle, which just fits the entrance holes of the beryllium shells, is mounted on the cradle. The beryllium sample is assembled, shell by shell, upon the cradle, with the entrance holes fitted down over the spindle. Figure 5 shows the cradle with the large sample half assembled. When the assembly is complete, the sample holder, which we have named the bird cage, is clamped down over the beryllium. The bird cage consists of a cylindrical aluminum base, upon which the sample will sit in the sample chamber of the manganese bath, and a close-fitting spider arrangement, whose four arms fit down around the beryllium and hook into slots in the base. Figure 6 shows sample Be #4 in the bird cage, assembled on the cradle. The tilting feature of the cradle has been utilized, and the sample is rotated to a vertical position. The sample is then released from the cradle and lifted by a forklift to a drum dolly on the working platform above the manganese bath. A slow-speed half-ton hoist lifts the sample from the dolly and lowers it into the sample chamber of the bath. Figure 7 shows the sample safely installed in the sample chamber.

TABLE 3. COMPOSITION OF BERYLLIUM SAMPLES^a

Component	Weight Per Cent
Be	98.70
BeO	1.25
C	0.123
Al	0.000570
Cr	0.000090
Fe	0.001345
Mg	0.000195
Mn	0.000100
N	0.000520
Ni	0.000145
Ti	0.000015
Ag	< 0.000003
Ca	< 0.000020
Co	0.000006
Cu	0.000045
Mo	< 0.000020
Pb	< 0.000020
Sc	0.000007
Si	0.000540
U	0.000055
W	< 0.000100
Zn	< 0.000010

a. Based on analysis of shell L-8.

Figure 5. Sample Cradle with Beryllium Sample Half Assembled.

DATA HANDLING

Data from two manganese bath detectors, the associated α -particle detector, a current integrator, a paraffin-moderated neutron detector, and a 5 cm x 5 cm NE-213 fast-neutron scintillator are routinely collected in Camac scalars. The latter two detectors monitored the neutron leakage from the bath. The temperatures indicated by two thermocouples mounted on the drift tube of the neutron generator are also recorded. An IBM PC/AT computer controls the collection of data. The MANSAT⁹ program performs the data analysis.

MANSAT is based upon the concept of manganese flow counting developed at the University of Michigan by D. M. Gilliam and others¹². The total ⁵⁶Mn activity in the system is considered to consist of two components. The bulk of the activity has been present for at least the homogenization time, τ , and is counted

Figure 6. Sample Be #4 Assembled on Cradle with Bird Cage.

with the an efficiency ϵ , which is the efficiency for detecting the calibrated ^{56}Mn activity mixed into the bath at calibration time. The second component is activity that has been too recently produced to allow full homogenization. It is counted with an effective efficiency of $\epsilon(1 - \delta)$, where δ represents the fractional efficiency loss for the activity that is not homogenized. The two mixing parameters, τ and δ , are not independent of each other. An approximate value for τ is adopted, and δ is adjusted to achieve consistency in the calculations of saturated activity. If the value of δ is too low, the calculated saturated activity values are relatively low at the beginning of the irradiation (the growth phase), gradually increasing to a constant value as the irradiation continues, and leveling off at a value below the activities calculated from the

Figure 7. Beryllium Sample in Place in Manganese Bath

decay phase of the experiment. If the value of δ is too high, the calculated saturated activity values begin high, decreasing with time and leveling off at a value higher than the values from the decay phase. MANSAT includes a search routine to vary δ until the slope of the calculated saturated activity curve is less than 10^{-7} . When the slope is thus minimized, the growth data are found to agree closely with the decay data.

A test of the homogenization time was conducted in conjunction with a neutron leakage measurement. With the pumps off, the bath was irradiated for two hours, during which time the detectors measured background activity. Then the two pumps were turned on simultaneously, and the activity measured in 30-second intervals. The rise in activity is represented in Figure 8. The solution is well homogenized within the 600-second homogenization time adopted for all manganese bath data reduction.

Figure 8. Mixing Test of Manganese Bath Pumps.

Manganese Bath Response Equations: ^{252}Cf Source

For a ^{252}Cf source the saturated activity, as indicated by the data from the n th counting period during the irradiation (growth phase), is given by

$$A_{sgn} = C_n A_{1n} / (A_{2n} - \delta A_{3n}), \quad (1)$$

with

$$A_{1n} = \exp(\lambda_s t_n) (\lambda_m - \lambda_s) / \lambda_m, \quad (2)$$

$$A_{3n} = \{1 - \exp[(\lambda_s - \lambda_m) \tau]\} [1 - \exp(-\lambda_s t_c)] / \lambda_s. \quad (4)$$

$$A_{2n} = \exp [(\lambda_s - \lambda_m) t_n] [\exp (-\lambda_m t_c) - 1] / \lambda_m + [1 - \exp (\lambda_s t_c)] / \lambda_s , \quad (3)$$

Following the removal of the ^{252}Cf source the saturated activity as indicated by the n th decay-phase count is given by

$$A_{sdn} = \frac{C_n (\lambda_m - \lambda_s) \exp [\lambda_m (t_n - t_I)]}{[1 - \exp (-\lambda_m t_c)] [\exp (-\lambda_s t_I) - \exp (-\lambda_m t_I)]} . \quad (5)$$

In Equations 1 to 5 C_n represents the total number of counts, corrected for dead-time losses, background, and residual activity, observed during the counting period t_c , which began at a time t_n after insertion of the source into the bath. The duration of the irradiation is given by t_I . The decay constants of manganese and the source are represented by λ_m and λ_s , respectively. The two mixing parameters, τ and δ , are described above.

The decay factors for ^{252}Cf contained in Equations 1 to 5 account for the effects of decay of the source during the irradiation. A further decay correction is applied to deduce the source strength at an earlier reference date, following determination of the weighted average of the saturated activity values.

Manganese Bath Response Equations: Neutron Generator

The response equations in the case of a ^{252}Cf source are relatively simple because the neutron source strength changes only slowly and predictably over the period of the irradiation. The neutron generator is less predictable. The source strength increases as the machine warms up and decreases as the target becomes depleted of its tritium. In addition there may be fluctuations in the performance of the ion source. These variations affect the ^{56}Mn counting in two ways. In the first place, variation of the neutron source strength requires that each count be normalized to the counting rate observed in the associated α -particle detector. In the second place, consideration must be given to the fact

that each ^{56}Mn data point contains contributions from activations produced during the whole prior history of the irradiation. To account for these effects, we developed a "neutron generator" version of MANSAT. It assumes that the source strength is constant over each counting period, the counting period being chosen to make this approximately so. The irradiation period is assumed to consist of a series of short irradiation periods, with the n th count containing decay-weighted contributions from all of the previous irradiation period. The homogenization time is presumed to extend back into the $n-1$ th counting period. Under these assumptions the manganese bath response takes the following form.

$$W_{sgn} = C_n / (G_{1n} - \delta G_{2n}) \quad , \quad (6)$$

with

$$G_{1n} = D_1 \sum_{k=1}^{n-1} \rho_{\alpha k} \exp [- (n - k - 1) \lambda t_c] + D_2 \rho_{\alpha n} \quad , \quad (7)$$

$$G_{2n} = D_3 \rho_{\alpha(n-2)} + D_1 \rho_{\alpha(n-1)} + D_2 \rho_{\alpha n} \quad . \quad (8)$$

The D's are combinations of decay parameters:

$$D_1 = [1 - \exp(-\lambda_m t_c)]^2 / \lambda_m \quad , \quad (9)$$

$$D_2 = t_c - [1 - \exp(-\lambda_m t_c)] / \lambda_m \quad , \quad (10)$$

$$D_3 = \exp(-\lambda_m t_c) [1 - \exp(-\lambda_m t_c)] / \lambda_m - t_c \exp(-\lambda_m \tau) \quad . \quad (11)$$

W_{sgn} is the saturation value of the ^{56}Mn activity, as indicated by the n th count observed during the growth period of the irradiation, normalized to the

decay-weighted associated particle counting rate. ρ_{an} is the average counting rate of the associated particle detector observed during the n th counting period. The irradiation continues for N counting periods, after which counting continues during the decay phase of the run. During the decay phase the α -normalized saturation activity is given by

$$W_{sdn} = \frac{C_n \lambda_m \exp[\lambda_m(t_n - t_1)]}{[1 - \exp(-\lambda_m t_c)]^2 \sum_{k=1}^N \rho_{ak} \exp[-(N-k)\lambda_m t_c]} \quad (12)$$

As with the neutron source version of the program, the mixing parameters τ and δ are determined by assuming a value for τ and adjusting δ to force the growth curve to be flat. The mixing parameters for the neutron source and neutron generator versions of MANSAT are very nearly equal, as they depend principally upon the mechanical characteristics of the manganese bath. There appears to be a very small dependence upon the neutron spectrum, and the value of δ is slightly smaller for the 14-MeV neutrons than for ^{252}Cf neutrons. In the present experiment we used a value of $\tau = 600$ sec. The fitted values of δ are about 0.19 for 14-MeV neutrons and 0.21 for ^{252}Cf neutrons.

Because both the neutron generator and ^{252}Cf source produce excellent statistical precision in the decay phase of the experiment, we use only the decay data. However, the growth data were also routinely calculated and examined for indications of any irregularities in the irradiation. Saturation values from the entire growth-decay cycle were plotted for examination. A typical plot is shown in Figure 9. Data from the growth phase are plotted as $\underline{1}$'s, and the decay data are indicated by $\underline{2}$'s. The facts that the history is very flat, and that the growth and decay data merge smoothly, indicate that the mixing parameters have been properly determined and that there are no serious experimental problems with this irradiation.

The statistically-weighted average of the W_{sdn} values is W_s^i for an irradiation using beryllium sample i , and W_s^o for an open beam irradiation. The

ratio W_s^i/W_s^o is the apparent multiplication M_A^i for the beryllium sample. M_A^i is the quantity that can be compared directly with calculation to test the effectiveness of cross section data and calculational methods. For the true multiplication of the sample the effects of the manganese bath upon the measurement must be considered.

MANGANESE BATH CHEMISTRY

In order to set up the MCNP model properly, and to be sure that there are no substantial amounts of absorbers in the $MnSO_4$ solution that would disturb the neutron economy of the system, it is necessary that the solution be well characterized. There is a certain amount of evaporation with time, and it is necessary to have a record of the consequent change in concentration to make appropriate corrections in the experimental data. For these reasons we measured the solution concentration routinely. Concentration measurements were made both gravimetrically (by evaporating a sample of solution to dryness and weighing the residue) and titrametrically (by titration with EDTA). Both methods are used, because earlier work⁹ showed that the level of impurities in the solution is indicated by the difference between the two measurements. For a solution of high purity the titrametric result gives the concentration of manganese, while the gravimetric method yields the total quantity of solids in the solution. Concentrations for the solution sample collected 2 June 1989 are shown in Part I of Table 4. Because the MCNP model of the experiment requires information concerning the number density of all component nuclei, the solution density is also measured. The density of the 2 June 1989 sample is shown in Table 4 along with the other concentration data.

TABLE 4. COMPOSITION OF THE MANGANESE BATH

I. Manganese Bath Concentration on 2 June 1989						
Gravimetric (g/Kg)		Titrametric (g/kg)		Ratio G/T	Density (g/ml)	
212.71		211.953		1.00357	1.23354	

II. Impurities in the Manganese Bath						
Element	At. Wt.	σ_{th} (barns)	Chem. Form	Concentration ($\mu\text{g/ml}$)		Σ (cm^{-1})
				Element	Sulfate	
Li	6.941	71	Li_2SO_4	0.045	0.358	2.772E-07
B	10.811	765	H_3BO_3	0.054	0.309	2.301E-06
Cd	112.41	2450	CdSO_4	0.093	0.172	1.221E-06
Fe	55.847	2.56	Fe_2SO_4	4.447	15.92	1.228E-07
Mg	24.305	0.063	MgSO_4	6.084	30.11	9.496E-09
Al	26.9815	0.233	$\text{Al}_2(\text{SO}_4)_3$	18.908	119.7	9.833E-08
Na	22.9898	0.53	Na_2SO_4	79	487.7	1.097E-06
Absorption associated with oxygen and sulfur in impurities						1.780E-06
Total absorption in impurities						6.906E-06
Ratio $\Sigma_{\text{Imp}} / \Sigma_{\text{Mn}}$						0.000494

The concentration history showed that throughout the course of the experiment the manganese concentration increased at an average rate of 0.036% per day. The experimental data were corrected for this concentration change.

To complete the characterization of the solution we have determined the level of various impurities in the bath. The impurities are summarized in Part II of Table 4. With the exception of boron, the impurity levels were measured by Inductively Coupled Plasma (ICP) analysis. For boron we use the results of

a colorimetric measurement. The right-hand column shows the macroscopic thermal neutron absorption cross section represented by each impurity element. To these values must be added the contributions from the sulfur and oxygen in the impurity sulfate molecule (or borate, in the case of boron). The total impurity macroscopic absorption cross section is approximately 0.05% of that of the manganese in the bath.

The list of impurities in Table 4 represents a somewhat abbreviated list, the list having been shortened somewhat to simplify the MCNP setup. A few other impurities, of negligible absorption, have been neglected.

MANGANESE BATH SYSTEMATIC EFFECTS

With the sample chamber empty, the saturated ^{56}Mn activity of the bath is given by the relation

$$W_s^o = \frac{4\pi}{\omega_\alpha} R_\alpha e f^o (1 - L^o - D^o - P^o - S^o + N_s) \quad (13)$$

With a beryllium sample in the chamber, the α -normalized saturated activity becomes

$$W_s^i = M_{\text{pure}}^i \frac{4\pi}{\omega_\alpha} R_\alpha e f^i \left[1 + \frac{N_s^i + N_{\text{Im}}^i + N_{\text{Be}}'^i - A_{\text{Be}}^i - A_{\text{Im}}^i - L^i - D^i - S^i - P^i}{1 + N_{\text{Be}}^i - A_{\text{Be}}^i} \right] \quad (14)$$

In Equations 2 and 3 the superscript o refers to the value of the particular quantity when the sample chamber is empty, and the superscript i refers to the value when the sample chamber contains beryllium sample i . ω_α is the solid angle subtended by the α detector, and R_α is the anisotropy correction factor required to relate the alpha counting rate to the total neutron yield. The efficiency factor e is the efficiency of the NaI(Tl) detectors for counting ^{56}Mn activity anywhere in the bath system. M_{pure} is the multiplication in just the beryllium of the sample. The factors N account for small multiplication contributions due to $(n,2n)$ reactions on the structural materials, including the manganese of the bath (N_s), impurities in the beryllium (N_{Im}), and neutrons returning from the bath to the beryllium (N_{Be}'). The factors A represent the

absorption of primary neutrons in the beryllium (A_{Be}^i), the absorption of returned neutrons in the beryllium (A_{Be}^r), and absorption of neutrons by the impurities in the beryllium (A_{Im}^i). The factors f represent the fraction of neutrons absorbed in manganese in the Maxwellian part of the spectrum in the bath, expressed by the relation

$$f = \frac{N_{Mn} \sigma_{Mn} (1 + Grs)}{N_{Mn} \sigma_{Mn} (1 + Grs) + N_H \sigma_H + N_O \sigma_O + N_S \sigma_S + \sum N_I \sigma_I} \quad (15)$$

In Eq. (15) the N 's are the number densities, and σ 's are the 2200 m/sec cross sections of manganese, hydrogen, oxygen, sulfur, and whatever impurities are in the bath. The term $(1 + Grs)$ is a correction, using the Westcott formalism, for the absorption in the manganese resonances, principally the one at 336 eV. The Westcott g factor is unity. The G represents a correction for self-shielding in the manganese resonances, and r represents Westcott's r factor, which is essentially the resonance integral. The resonance correction depends upon the spectrum, which in our case varies with the amount of beryllium between the source and the bath. The resonance correction is not large, and the ratio f^0/f^i varies very little from unity. L , D , S , and P are corrections for leakage, duct streaming, absorption in structural materials, and absorption in high-energy charged particle reactions, respectively. Most of the corrections are spectrum-dependent. With no beryllium sample present, the manganese bath sees the pristine 14-MeV spectrum, which becomes thermalized in the bath. When beryllium is present, it acts as a moderator as well as a multiplier, and the manganese bath sees a partially moderated spectrum. The systematic effects have different values for each sample thickness.

Equations (13) and (14) can be combined to deduce the true multiplication value for the beryllium sample:

$$M_{pure}^i = \frac{W_s^i f^o}{W_s^o f^i} \frac{1 - L^o - D^o - S^o - P^o + N_s^o}{\left[1 + \frac{N_s^i + N_{Im}^i + N_{Be}^i - A_{Be}^i - A_{Im}^i - L^i - D^i - S^i - P^i}{1 + N_{Be}^i - A_{Be}^i} \right]} \quad (16)$$

Equation 16 shows an impressive array of corrections to be evaluated. Fortunately most of the effects are small in size. For fission, (α ,n), and (γ ,n) sources the manganese bath systematic effects have been widely studied by both experimental and calculational techniques, and corrections can be made to a high level of confidence. For 14-MeV neutrons some of the corrections are much larger than for lower-energy sources, and their uncertainties are also larger. Values of the corrections were extracted from detailed edits of the Monte Carlo calculation of the experiment.

CALCULATIONAL MODEL

The experiment was modeled by Dr. Wiley Davidson of Los Alamos¹³ for calculations using the Monte Carlo code MCNP. An extremely detailed model of the experiment was constructed. Figure 10 is a diagram of the model, as generated by the MCNP program. Cross sections used were from ENDF/B-V, with the exception of beryllium, for which two different cross-section sets were used. The first calculations were made using the evaluation by Young and Stewart (YS),¹⁴ because the ENDF/B-VI data for beryllium were not yet available to MCNP. The purpose of Young and Stewart in performing their evaluation was to cast an approximation to the LLL evaluation into the ENDF/B-V format. It was not accepted for ENDF/B-V, but is available from ENDF/A, as well as the European Fusion File. The LLL evaluation, with some further modifications, became the ENDF/B-VI data file. Recently this program supported the necessary conversion of MCNP to utilize the ENDF/B-VI beryllium cross section data. A description of the conversion work appears in the Appendix.

A large variety of MCNP tallies were kept to allow extraction of the values of individual corrections as well as the value of the apparent

Figure 10. MCNP-generated diagram of the calculational model. Be#3 is represented.

multiplication M_A . The values of the systematic effects for the open beam and the individual beryllium samples are listed in Table 5, for calculations using the YS beryllium data, and Table 6, for calculations using the ENDF/B-VI beryllium data. The manganese bath corrections are relatively insensitive to the particular beryllium data set used. The differences between the two evaluations is registered in the (n,2n) and absorption events recorded for

each. The uncertainties shown are the MCNP statistical uncertainties, except for the parasitic absorption correction P; which carries a 20% uncertainty because of the uncertainties in the cross sections.

When a neutron returned from the bath to the beryllium sample, it was flagged by MCNP. The indirect multiplication effect was evaluated from multiplications due to the flagged neutrons. The flagged tallies also showed non-multiplying absorptions of returned neutrons. Absorptions in beryllium include not only (n,γ) events but also (n,α) and (n,t) reactions, which are non-negligible. In Table 5 the effects labeled N'_{Be} and A'_{Be} represent the effects of $(n,2n)$ and absorption reactions, respectively, due to neutrons returning from the bath to the beryllium. Multiplication due to the returned neutrons is very low, because neutrons undergoing enough scattering events to return them to the beryllium will most probably lose enough energy to take them below the $(n,2n)$ threshold.

TABLE 5. MANGANESE BATH CORRECTIONS: YOUNG-STEWART BERYLLIUM DATA

Correction	Open	Be #1	Be #2	Be #3	Be #4
f^o/f^i	-----	1.0000 $\pm$ 0.0025	1.0017 $\pm$ 0.0025	1.0027 $\pm$ 0.0025	1.0039 $\pm$ 0.0025
L: Leakage	0.0192 $\pm$ 0.0016	0.0143 $\pm$ 0.0014	0.0087 $\pm$ 0.0010	0.0067 $\pm$ 0.0008	0.0047 $\pm$ 0.0011
D: Duct strm	0.0006 $\pm$ 0.0002	0.0006 $\pm$ 0.0002	0.0009 $\pm$ 0.0002	0.0010 $\pm$ 0.0002	0.0010 $\pm$ 0.0003
S: Struct Abs	0.0120 $\pm$ 0.0001	0.0179 $\pm$ 0.0002	0.0349 $\pm$ 0.0002	0.0444 $\pm$ 0.0003	0.0531 $\pm$ 0.0005
P: Par Abs	0.1332 $\pm$ 0.0267	0.1012 $\pm$ 0.0202	0.0586 $\pm$ 0.0117	0.0453 $\pm$ 0.0091	0.0318 $\pm$ 0.0064
N _S : Struct Mult.	0.0224 $\pm$ 0.0001	0.0187 $\pm$ 0.0001	0.0152 $\pm$ 0.0001	0.0143 $\pm$ 0.0001	0.0137 $\pm$ 0.0002
N _{Im} : Be Imp Abs	0.0000 $\pm$ 0.0000	0.0001 $\pm$ 0.0001	0.0001 $\pm$ 0.0001	0.0001 $\pm$ 0.0001	0.0002 $\pm$ 0.0001
N' _{Be} : Ret (n,2n)	0.0000 $\pm$ 0.0000	0.0023 $\pm$ 0.0003	0.0049 $\pm$ 0.0005	0.0055 $\pm$ 0.0005	0.0067 $\pm$ 0.0009
A' _{Be} : Ret abs Be	0.0000 $\pm$ 0.0000	0.0013 $\pm$ 0.0001	0.0124 $\pm$ 0.0002	0.0254 $\pm$ 0.0003	0.0482 $\pm$ 0.0007
A _{Im} : Abs Be Imp.	0.0000 $\pm$ 0.0000	0.0008 $\pm$ 0.0001	0.0083 $\pm$ 0.0001	0.0185 $\pm$ 0.0001	0.0380 $\pm$ 0.0003
N _{Be} : Be (n,2n)	0.0000 $\pm$ 0.0000	0.3341 $\pm$ 0.0012	0.8407 $\pm$ 0.0031	1.0285 $\pm$ 0.0037	1.2086 $\pm$ 0.0043
A _{Be} : Be abs	0.0000 $\pm$ 0.0000	0.0303 $\pm$ 0.0001	0.1022 $\pm$ 0.0005	0.1555 $\pm$ 0.0006	0.2448 $\pm$ 0.0009

TABLE 6. MANGANESE BATH CORRECTIONS: ENDF/B-VI BERYLLIUM DATA

Correction	Open	Be #1	Be #2	Be #3	Be #4
f^o/f^i	-----	1.0001 $\pm$ 0.0060	1.0017 $\pm$ 0.0053	1.0028 $\pm$ 0.0056	1.0040 $\pm$ 0.0054
L: Leakage	0.0202 $\pm$ 0.0012	0.0154 $\pm$ 0.0010	0.0090 $\pm$ 0.0007	0.0064 $\pm$ 0.0007	0.0045 $\pm$ 0.0006
D: Duct strm	0.0006 $\pm$ 0.0001	0.0008 $\pm$ 0.0001	0.0009 $\pm$ 0.0001	0.0009 $\pm$ 0.0001	0.0009 $\pm$ 0.0001
S: Struct Abs	0.0120 $\pm$ 0.0001	0.0179 $\pm$ 0.0001	0.0349 $\pm$ 0.0002	0.0436 $\pm$ 0.0002	0.0535 $\pm$ 0.0002
P: Par Abs	0.1333 $\pm$ 0.0005	0.1037 $\pm$ 0.0005	0.0626 $\pm$ 0.0004	0.0488 $\pm$ 0.0004	0.0341 $\pm$ 0.0003
N _S : Struct Mult.	0.0225 $\pm$ 0.0001	0.0190 $\pm$ 0.0001	0.0155 $\pm$ 0.0001	0.0146 $\pm$ 0.0001	0.0136 $\pm$ 0.0001
N _{Im} : Be Imp Abs	0.0000 $\pm$ 0.0000	0.0001 $\pm$ 0.0001	0.0001 $\pm$ 0.0001	0.0001 $\pm$ 0.0001	0.0002 $\pm$ 0.0001
N' _{Be} : Ret (n,2n)	0.0000 $\pm$ 0.0000	0.0020 $\pm$ 0.0002	0.0052 $\pm$ 0.0003	0.0054 $\pm$ 0.0003	0.0067 $\pm$ 0.0003
A' _{Be} : Ret abs Be	0.0000 $\pm$ 0.0000	0.0011 $\pm$ 0.0001	0.0111 $\pm$ 0.0001	0.0227 $\pm$ 0.0002	0.0445 $\pm$ 0.0003
A _{Im} : Abs Be Imp.	0.0000 $\pm$ 0.0000	0.0008 $\pm$ 0.0001	0.0085 $\pm$ 0.0001	0.0186 $\pm$ 0.0001	0.0389 $\pm$ 0.0002
N _{Be} : Be (n,2n)	0.0000 $\pm$ 0.0000	0.3137 $\pm$ 0.0010	0.8070 $\pm$ 0.0027	0.9915 $\pm$ 0.0031	1.1732 $\pm$ 0.0036
A _{Be} : Be abs	0.0000 $\pm$ 0.0000	0.0258 $\pm$ 0.0002	0.0919 $\pm$ 0.0005	0.1411 $\pm$ 0.0007	0.2256 $\pm$ 0.0010

EXPERIMENTAL TESTS OF SYSTEMATIC EFFECTS

It was possible to obtain experimental values for three of the corrections calculated by MCNP. These were the parasitic absorption, leakage, and structural absorption for the open beam, and the structural absorption for samples Be #2 and Be #4.

The correction for parasitic absorption is the largest and most troublesome of the corrections. Estimates of its magnitude have ranged as high as 25%.¹⁵ The presence of the associated particle detector makes it possible to obtain an experimental check upon the value of this correction, which normally can only be calculated. The efficiencies of the manganese bath counters were measured by adding a calibrated aliquot of ⁵⁶Mn activity to the bath. The α -detector subtends a fractional solid angle ($\omega_\alpha/4\pi$) of 1.5816×10^{-5} . The anisotropy factor R_α was calculated using the computer program RALPHA.¹⁶ The remaining correction factors contained in Equation (13) are small and have small uncertainties. The values of those parameters not listed in Tables 5 and 6 are shown in Table 7. The resultant value for the

TABLE 7. EXTRACTION OF PARASITIC ABSORPTION CORRECTION

Parameter	Value, Ctr #1	Value, Ctr #2
W_s°	5.42667 ± 0.00036	5.34808 ± 0.00098
$\omega_\alpha / 4\pi$	$(1.5816 \pm 0.0035)E-05$	$(1.5816 \pm 0.0035)E-05$
e	$(2.2867 \pm 0.0077)E-04$	$(2.2538 \pm 0.0076)E-04$
R_α	1.146 ± 0.018	1.146 ± 0.018
f°	0.39224 ± 0.00098	0.39224 ± 0.00098
P	0.136 ± 0.014	0.136 ± 0.014

parasitic absorption correction deduced from this approach is 0.136. The value calculated by MCNP is 0.133. The latter value is higher than the value (0.123) previously reported,¹⁷ because the final MCNP calculations included absorption by the (n,d), (n,t), and (n,³He) reactions in the bath. For 14-MeV neutrons these reactions become significant factors, whereas for lower energy neutrons only the (n,p) and (n, α) cross sections are normally considered.

The recent ENDF/B-VI evaluation for oxygen¹⁸ raised the ¹⁶O(n, α)¹³C cross sections by approximately 25% above the ENDF/B-V values. As our experimental result supports the ENDF/B-V value, we continue to use the latter cross sections.

The leakage of the bath was estimated from a transmission measurement. The counting rate of a modified "long" counter was observed with the bath empty and then filled with the solution. A value of 0.026 was obtained. The agreement with the calculated value of 0.020 is satisfactory in view of the complexity of the bath geometry and the fact that the transmission measurement was made at only one position.

The structural absorption correction was measured by repeating the experiment, both open and sample irradiations, with an added aluminum layer around the sample chamber. The added aluminum doubled the absorption effect. The drift tube and sample support could not be doubled in this way because of space limitations, and sample-in measurements were made for only samples Be #2 and Be #4. Comparisons of the measurements with theoretical results from the MCNP calculations are shown in Table 8. Agreement is satisfactory in view of the uncertainties involved. These comparisons represent all the cases for which experimental tests could be made.

TABLE 8. MEASURED SYSTEMATIC EFFECTS

Effect	Experiment	MCNP Calculation
Leakage (no sample)	0.026 ± 0.003	0.0203 ± 0.002
Abs in Al (open)	0.007 ± 0.001	0.008 ± 0.001
Abs in Al (Be #2)	0.016 ± 0.002	0.014 ± 0.001
Abs in Al (Be #4)	0.026 ± 0.003	0.020 ± 0.001
Parasitic Abs (no sample)	0.136 ± 0.014	0.133 ± 0.025

EXPERIMENTAL RESULTS

In the experimental program twenty-two irradiations were made with the open beam, three with Be #1, 5 with Be #2, four with Be #3, and eleven with Be #4. With the aluminum structure doubled, four irradiations were made with the open beam, two with Be #2, and four with Be #4. The alpha-normalized saturated activities of these runs, together with their statistical standard errors, are listed in Table 9. The data have been corrected for the effects of concentration increase (maximum 0.6% change) and (d,d) neutron contamination from deuterium deposited in the target (maximum 0.2% change). The latter effect was determined from a series of blank-target irradiations, in which the ^{56}Mn activity was normalized to the counting rate from the current integrator, rather than from the α -particle detector.

TABLE 9. EXPERIMENTAL DATA

Date	W_s , Ctr #1	W_s , Ctr #2
I. Open-Beam Runs		
30 Oct 1989	5.36722 $\pm$ 0.00054	5.29719 $\pm$ 0.00053
1 Nov 1989	5.35748 $\pm$ 0.00066	5.28633 $\pm$ 0.00066
3 Nov 1989	5.35629 $\pm$ 0.00066	5.28408 $\pm$ 0.00066
14 Nov 1989	5.35636 $\pm$ 0.00065	5.28422 $\pm$ 0.00064
15 Nov 1989	5.35573 $\pm$ 0.00057	5.28408 $\pm$ 0.00057
4 Dec 1989	5.35454 $\pm$ 0.00055	5.28381 $\pm$ 0.00055
5 Dec 1989	5.35370 $\pm$ 0.00057	5.28233 $\pm$ 0.00057
14 Dec 1989	5.35953 $\pm$ 0.00059	5.27595 $\pm$ 0.00059
18 Dec 1989	5.33631 $\pm$ 0.00060	5.26441 $\pm$ 0.00059
19 Dec 1989	5.33591 $\pm$ 0.00062	5.26298 $\pm$ 0.00062
21 Dec 1989	5.34051 $\pm$ 0.00062	5.26740 $\pm$ 0.00062
5 Jan 1990	5.34414 $\pm$ 0.00054	5.27204 $\pm$ 0.00053
8 Jan 1990	5.34491 $\pm$ 0.00055	5.27184 $\pm$ 0.00055
9 Jan 1990	5.33665 $\pm$ 0.00082	5.26298 $\pm$ 0.00081
23 Jan 1990	5.35432 $\pm$ 0.00055	5.27629 $\pm$ 0.00055
24 Jan 1990	5.35452 $\pm$ 0.00057	5.27705 $\pm$ 0.00057
28 Mar 1990	5.36082 $\pm$ 0.00061	5.28208 $\pm$ 0.00060
29 Mar 1990	5.36071 $\pm$ 0.00066	5.28329 $\pm$ 0.00065
30 Mar 1990	5.35945 $\pm$ 0.00063	5.28202 $\pm$ 0.00063
15 May 1990	5.37620 $\pm$ 0.00057	5.30112 $\pm$ 0.00057
16 May 1990	5.37403 $\pm$ 0.00055	5.29677 $\pm$ 0.00054
17 May 1990	5.37503 $\pm$ 0.00057	5.29823 $\pm$ 0.00057
II. Be #1 Runs		
17 Nov 1989	7.44449 $\pm$ 0.00071	7.34627 $\pm$ 0.00071
20 Nov 1989	7.44366 $\pm$ 0.00075	7.34254 $\pm$ 0.00075
21 Nov 1989	7.44232 $\pm$ 0.00069	7.34291 $\pm$ 0.00068
II. Be #2 Runs		
7 Nov 1989	10.33450 $\pm$ 0.00097	10.19778 $\pm$ 0.00097
8 Nov 1989	10.32974 $\pm$ 0.00133	10.19456 $\pm$ 0.00132
9 Nov 1989	10.33963 $\pm$ 0.00087	10.19963 $\pm$ 0.00087
10 May 1990	10.34111 $\pm$ 0.00075	10.19493 $\pm$ 0.00074
11 May 1990	10.34372 $\pm$ 0.00088	10.19709 $\pm$ 0.00087

TABLE 9. EXPERIMENTAL DATA

Date	W_s , Ctr #1	W_s , Ctr #2
IV. Be #3 Runs		
7 Dec 1989	11.07342 ± 0.00092	10.92469 ± 0.00091
8 Dec 1989	11.07290 ± 0.00084	10.92640 ± 0.00083
11 Dec 1989	11.07103 ± 0.00088	10.92236 ± 0.00087
12 Dec 1989	11.07612 ± 0.00085	10.93125 ± 0.00086
V. Be #4 Runs		
28 Dec 1989	11.37190 ± 0.00102	11.21725 ± 0.00101
29 Dec 1989	11.37035 ± 0.00090	11.21506 ± 0.00089
3 Jan 1990	11.37221 ± 0.00098	11.21667 ± 0.00098
15 Jan 1990	11.36733 ± 0.00088	11.21010 ± 0.00087
16 Jan 1990	11.36300 ± 0.00100	11.21138 ± 0.00099
18 Jan 1990	11.36715 ± 0.00103	11.20896 ± 0.00102
19 Jan 1990	11.36902 ± 0.00091	11.20811 ± 0.00090
3 Apr 1990	11.39469 ± 0.00094	11.23329 ± 0.00093
23 May 1990	11.41276 ± 0.00074	11.25153 ± 0.00073
24 May 1990	11.41349 ± 0.00101	11.24946 ± 0.00101
25 May 1990	11.41076 ± 0.00077	11.24860 ± 0.00077
VI. Open Beam with Extra Aluminum		
20 Apr 1990	5.32667 ± 0.00060	5.25058 ± 0.00059
23 Apr 1990	5.33454 ± 0.00065	5.25924 ± 0.00064
24 Apr 1990	5.33318 ± 0.00066	5.25636 ± 0.00065
25 Apr 1990	5.33290 ± 0.00062	5.25570 ± 0.00062
VII. Be #2 with Extra Aluminum		
7 May 1990	10.17227 ± 0.00094	10.02786 ± 0.00094
8 May 1990	10.19174 ± 0.00073	10.04929 ± 0.00072
VIII. Be #4, With Extra Aluminum		
27 Apr 1990	11.11095 ± 0.00094	10.94972 ± 0.00094
30 Apr 1990	11.12775 ± 0.00098	10.96898 ± 0.00098
1 May 1990	11.12437 ± 0.00086	10.96983 ± 0.00085
3 May 1990	11.12066 ± 0.00088	10.96409 ± 0.00087

From the experimental data of Table 9 the raw, or apparent, values of the multiplication were calculated. The results are summarized in Table 10. The measured apparent multiplication values are just the ratios of ^{56}Mn α -normalized saturated activities induced in the bath with and without the sample in place. No corrections for systematic effects have been made at this point. The quoted errors represent just the standard deviations of the replications of the experiment. The calculated values in Table 10 are the MCNP results.¹³ These values are the ratios of tallied capture events in the manganese in the bath. It is just the capture events that produce the ^{56}Mn activity that is counted in the experiment. The results are shown graphically in Figure 11. Agreement is excellent in all cases. The multiplication values calculated with the

TABLE 10 APPARENT MULTIPLICATION IN BULK BERYLLIUM

I. 14-MeV Neutrons					
Sample	Experiment	Calculation			
		Y-S Be	C_{YS}/E	ENDF/B-VI Be	C_{VI}/E
Be #1	1.390 ± 0.001	1.382 ± 0.009	0.994	1.360 ± 0.007	0.978
Be #2	1.928 ± 0.001	1.902 ± 0.011	0.987	1.870 ± 0.009	0.970
Be #3	2.072 ± 0.002	2.037 ± 0.012	0.983	2.013 ± 0.009	0.972
Be #4	2.127 ± 0.001	2.077 ± 0.015	0.976	2.087 ± 0.009	0.981

II. ^{252}Cf Source			
Sample	Experiment	Calc, Y-S Be	C_{YS}/E
Be #1	1.036 ± 0.002	1.038 ± 0.009	1.002
Be #2	1.020 ± 0.002	1.021 ± 0.009	1.001
Be #3	0.979 ± 0.002	0.987 ± 0.009	1.008
Be #4	0.906 ± 0.002	0.907 ± 0.008	1.001

ENDF/B-VI beryllium data are only two to three per cent lower than those calculated with the Young-Stewart data, even though the (n,2n) cross sections

are about 15% lower. This is because the absorption cross sections are also lower for the ENDF/B-VI data.

Figure 11. Apparent multiplication of 14-MeV neutrons in beryllium samples

Also shown in Table 10 are the raw multiplication values measured for a ^{252}Cf source. The irradiations using the ^{252}Cf source have a dual purpose. Since the manganese bath systematic effects have been studied in more detail for fission neutron sources than for any other neutron source, the ^{252}Cf model

serves as a benchmark test for the MCNP calculations. Further, since (n,2n) multiplication is much lower for a fission source than for a 14-MeV source, the ^{252}Cf irradiations display the effects of neutron absorption in beryllium. A measured multiplication value significantly lower than the calculated value could indicate that impurities are present in the beryllium in quantities higher than those expected. The close agreement between calculation and experiment in this case indicates that the beryllium impurities are correctly accounted for.

The true values for multiplication in beryllium are obtained by applying the corrections listed in Tables 5 and 6. The corrected multiplication values for the four Be samples are shown in Table 11. The values shown represent the net corrected multiplication in each sample, including the effects of neutrons returning from the bath to the beryllium. The net corrected values of multiplication in the bulk beryllium samples are shown graphically in Figure 12.

TABLE 11. NET CORRECTED MULTIPLICATION IN BULK BERYLLIUM, 14-MEV NEUTRONS

Sample	Experiment	Calculation			
		Y-S Be	C_{YS}/E	ENDF/B-VI Be	C_{VI}/E
Be #1	1.311 ± 0.035	1.304 ± 0.001	0.995	1.288 ± 0.001	0.982
Be #2	1.764 ± 0.043	1.738 ± 0.003	0.985	1.715 ± 0.003	0.972
Be #3	1.904 ± 0.046	1.873 ± 0.004	0.984	1.850 ± 0.003	0.972
Be #4	1.988 ± 0.048	1.964 ± 0.004	0.988	1.948 ± 0.004	0.980

Figure 12. Net corrected multiplication in beryllium.

CONCLUSIONS

The multiplication of 14-MeV neutrons in bulk beryllium has been measured for four thicknesses of beryllium, ranging from 4.6 cm to 19.9 cm. The measured values have uncertainties ranging from 2.4% to 2.7%, and are in excellent agreement with the predictions based upon microscopic data. Residual differences may be at least partially due to uncertainties in auxiliary data such as the $^{16}\text{O}(n, \alpha)^{13}\text{C}$ cross section. We see no evidence of a substantial calculation-experiment discrepancy such as that mentioned by Cloth et al.¹ and other experimenters. Indeed, whereas the nature of the supposed discrepancy was that measured values of the multiplication were lower than calculated, our measured values are slightly higher than those calculated using data from

either of the two beryllium evaluations considered. Agreement with experiment is slightly better for the Young-Stewart evaluation than for the ENDF/B-VI data. However, the two evaluations differ very little in net multiplication effect, and both agree well with our measurements. Either data set can be used with confidence in predicting the neutron multiplication of neutrons in fusion reactor blankets containing beryllium.

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APPENDIX

The following pages contain a description by R. C. Little and R. E. Seamon, of Los Alamos National Laboratory, of their conversion of the MCNP Monte Carlo code to accommodate the use of the ENDF/B-VI beryllium cross section data.

A Description of an ENDF/B-VI ⁹Be Data Library for MCNP
and
A Modified Version of MCNP to Use the New Data

by

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INTRODUCTION

The MCNP Monte Carlo radiation transport code (Ref. 1) has always taken advantage of the most modern evaluated nuclear data. Most recently, the primary source of evaluated nuclear data for MCNP has been ENDF/B-V (Ref. 2). Although the format (Ref. 3) for ENDF/B-V allowed correlated energy-angle distributions of secondary neutrons, those File 6 possibilities were never implemented or used. Only the implicit secondary energy-angle correlation for elastic and discrete-inelastic scattering was used. The secondary energy distributions and secondary angular distributions of neutrons from all other reactions (e.g., (n,2n) and fission) were always completely uncorrelated. This situation has changed significantly as a result of a more general format description referred to as ENDF-6 (Ref. 4) available for ENDF/B-VI.

The ENDF-6 format introduces a completely revised and expanded File 6 for evaluations. The purpose of File 6 is to allow representation of reaction products in energy and angle. In particular, evaluators are to use File 6 when the energy and angular distributions of the emitted particle must be coupled. The production cross section (in barns/steradian) for a given reaction product is

$$\sigma_i(\mu, E, E') = \sigma(E) y_i(E) f_i(\mu, E, E') / 2\pi$$

where i denotes the reaction product, E is the incident energy, E' is the energy of the product emitted with cosine μ , $\sigma(E)$ is the interaction cross section, y_i is the product yield or multiplicity, and f_i is the normalized distribution so that

$$\int dE' \int d\mu f_i(\mu, E, E') = 1 \quad .$$

Several different laws are available to describe $f_i(\mu, E, E')$ in File 6. One of the available laws (Law 7) is the laboratory angle-energy law. The data for Law 7 are sorted according to (E, μ, E') ; i.e., the secondary energy is a function of the secondary scattering angle. Both are functions of the incident energy.

The ENDF/B-VI evaluation of ^9Be (Ref 5) takes advantage of File 6-Law 7 to represent secondary neutrons from the $(n,2n)$ reaction. The objectives of the current work are to define a new MCNP data-library format for this Law 7, to process the ENDF/B-VI evaluation of ^9Be into that new data-library format, to modify MCNP to read and correctly sample from the Law 7 data, to validate the previous steps, and to make the package available to individuals at the Los Alamos National Laboratory for MCNP analysis of ongoing experiments.

NEW DATA-LIBRARY FORMAT

The format of MCNP data libraries is described in detail in Appendix F of Ref 1. Separate portions of the libraries exist for angular distributions of secondary neutrons (see pages 505-506 of Ref 1) and for energy distributions of secondary neutrons (see pages 506-513 of Ref. 1). As described in the Introduction, the secondary distributions represented on the libraries up to now have been uncorrelated. (The exceptions are elastic scattering and discrete-level inelastic scattering. In both cases, for a particular incident neutron energy, each secondary scattering cosine uniquely determines a secondary energy.) Obviously, the advent of ENDF-6 generalized formats requires new MCNP data-library formats as well.

The strategy employed to define an MCNP data-library format to represent Law 7 data is as follows: (1) Secondary neutron angular distributions are generated from the Law 7 data and provided in the AND Block (Table F.12 of Ref. 1). There is no change from the current procedures in which all evaluated angular distributions are processed into 32 equally-likely secondary cosine bins. This method is appropriate for Law 7, because secondary angular distributions are a function of incident energy only. (2) A new format is defined for the DLW Block (Table F.14 of Ref. 1). The new MCNP law is called Law 67 (for ENDF/B File 6-Law 7). The format for MCNP Law 67 is fully defined in Table I, which is similar to parts a-j of Table F.14 of Ref. 1.

PROCESSING OF ENDF/B-VI ^9Be

Generally, ENDF/B data are processed into a format appropriate for MCNP by the NJOY code (Ref. 6). In this section we describe a somewhat non-standard use of NJOY for the current problem.

The ENDF/B-VI evaluation of ^9Be was made available on the Common File System (CFS) at Los Alamos by R. E. MacFarlane, Group T-2, as MAT-425 on /ENDF/6/NEUTRON/BE/9. To use effectively the current production version of NJOY, the evaluation was re-cast in ENDF/B-V format. ENDF/B File 6 angular distribution data were translated into an identical, but more traditional, File 4 representation using the code GMG. For the purpose of the initial NJOY processing, the ENDF/B File 6 energy distribution data were replaced with "place-holder" File 5 data generated in code MAKD, designed to represent accurately the neutron energy deposition. The following modules of NJOY were then executed to produce a preliminary MCNP data library: MODER, RECONR, BROADR, HEATR, and ACER.

Using the utilities COPY and EDIT, the File 6-Law 7 data were incorporated into the preliminary MCNP ACE library created by NJOY. The

secondary energy distribution data were added in the format described in Table I. Additional information, such as average secondary energies, was obtained from the CKHOW code. It is anticipated that the functions of GMG, MAKD, and CKHOW will ultimately be added to NJOY as part of a comprehensive upgrade of NJOY and MCNP associated with ENDF/B-VI. (It should be understood that all this work has been done only for ^9Be , because File 6-Law 7 is used for this isotope and this isotope alone.) The final ENDF/B-VI ^9Be MCNP data library (in Type 3 ACE format) was stored on CFS as /076997/MYREDSTUFF/BEST1.

MCNP MODIFICATIONS

A patch was written to Version 4.1 of MCNP (described in Ref. 7) to allow the code to read and sample from the new MCNP Law 67 data described above. The patch modifies MCNP Subroutine ACECAS, which is the routine that samples the energy of a secondary particle from the MCNP law data. The patch is available on CFS as /MYREDSTUFF/BE9/PATCHBE9.

When the modified ACECAS recognizes the need to sample from MCNP Law 67, the routine first calls ACECOS to sample the secondary scattering cosine. Then the secondary energy is sampled as a function of the sampled secondary scattering cosine. If the sampled value of the secondary scattering cosine lies between two cosine values at which secondary energy distributions are tabulated, a random number is used to select one or the other data set to sample from. When the incident energy is between two energy values at which secondary distributions are tabulated (and histogram interpolation is not invoked), a scaling method is employed, similar to that described in Section VI of Ref. 8. A secondary energy is sampled from the data provided for both incident energies; the scaling formalism is then used to determine the actual secondary energy.

The patched executable version of MCNP (for CRAY Y-MPs operating under CTSS) is available on CFS as /MYREDSTUFF/BE9/MCNPBE9. Note that this code does not currently support the MCNP point detector or DXTRAN capabilities when used in conjunction with MCNP Law 67.

VALIDATION OF DATA LIBRARY / CODE PACKAGE

There were three major aspects of our validation of the data library / code package: (1) differential and integral testing of the "traditional" portions of the data library, (2) differential and integral testing of the new (i.e., Law 67) portion of the data library, and (3) testing of various aspects of the patched version of MCNP using the new ENDF/B-VI ^9Be data library. In each case, the goal of our validation effort was to ensure that the package reproduced the intentions of the evaluators. Each aspect of our validation effort will now be described briefly.

Differential and integral testing of the "traditional" (i.e., non-Law 67) portion of the data library was accomplished with the MRKACR, MARK (Ref. 9), XSFICHE (Ref. 10), and XDATAP (Ref. 11) codes. MRKACR and MARK were used to compare integral, group-averaged cross sections for each reaction from the MCNP library and the original ENDF library, respectively. XSFICHE provided interpreted microfiche listings of the data found on the MCNP cross-section

library. Finally, XDATAP was used to produce differential plots from the MCNP library and from the original ENDF library. The data testing efforts described in this paragraph led, in all cases, to satisfactory conclusions.

To ensure correctness of the Law 67 data on the MCNP library, a new code, INFO67, was written. The code reads the MCNP data library and provides an interpreted listing of all of the Law 67 data found. The results therefrom are voluminous for the current case, with triplets of data (energy, probability density function, and cumulative density function) for as many as 91 secondary energy points, each for 21 scattering cosines, each for 17 incident energies (from 2.01 MeV to 20.0 MeV). Despite the volume of data, the output from INFO67 has allowed us to compare data values found on the MCNP library with those from the original ENDF/B-VI File 6-Law 7 tabulation. In all cases compared, agreement was obtained.

The most demanding testing of the data library / MCNP combination was carried out using the patched version of MCNP with the ENDF/B-VI ^9Be data library. First, a significant amount of dynamic debugging was performed to check the flow of the code when sampling from Law 67 data in the modified ACECAS subroutine. In all cases, the code performed as expected. Second, a series of simple MCNP calculation were performed, in which the source energy in a pure ^9Be problem was E_n , and a neutron cutoff energy was set at $E_n - \epsilon$, with $\epsilon = 1.0 \times 10^{-6}$. The goal was to allow MCNP to repeatedly sample secondary energies from the (n,2n) reaction. By picking E_n equal to values of incident neutron energy at which secondary data were tabulated, it was possible to compare average energies sampled by MCNP to average energies computed from integrations of the actual ENDF/B-VI data. Results confirmed our conclusion that the ENDF/B-VI File 6-Law7 data have been correctly interpreted and sampled by MCNP.

AVAILABILITY OF DATA LIBRARY / CODE PACKAGE

The ^9Be data library and modified MCNP have been made available to J. W. Davidson of Group A-3 at Los Alamos. The procedure for using the package is as follows:

```
MASS GET/MYREDSTUFF/BE9/MCNPBE9 (executable MCNP)
MASS GET/MYREDSTUFF/BE9/XSDIR (updated directory file)
MCNPBE9 INP=...
```

The ZAID of the ENDF/B-VI ^9Be library is 4009.60C. Finally, the user should note that the (n,2n) reaction is given by MT=16. This information is required MCNP tallies using the FM feature.

SUMMARY

A new MCNP data-library format has been defined to allow correlated energy-angle data from ENDF-6 File 6-Law 7 evaluations. The ENDF/B-VI evaluation of ^9Be has been processed into that format. MCNP has been modified to correctly sample from the new data. The data library / code package has undergone significant testing to validate the work done to satisfy the tasks described above. The entire package has been made available to others at Los

Alamos to allow further MNCP analysis of earlier ^9Be multiplication experiments.

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Table I

Format Description of MCNP DLW Block Law 67

Law _i =67		Laboratory Energy-Angle Law (from ENDF-6 File 6 Law 7)	
<u>Location</u>	<u>Parameter</u>	<u>Description</u>	
LDAT(1)	NR	Number of interpolation regions	
LDAT(2)	NBT(I), I=1, NR	ENDF interpolation parameters. If NR=0, NBT and INT are omitted and linear-linear interpolation is used.	
LDAT(2+NR)	INT(I), I=1, NR		
LDAT(2+2*NR)	NE	Number of incident energies	
LDAT(3+2*NR)	EIN(I), I=1, NE	Incident energies	
LDAT(3+2*NR+NE)	LEIN(I), I=1, NE	Location of data for each incident energy (relative to JXS(11) or JXS(19))	
Data for EIN (1) (let K=3+2*NR+2*NE):			
LDAT(K)	INTMU	Interpolation parameter between secondary cosines (INTMU=1 is histogram, INTMU=2 is linear-linear)	
LDAT(K+1)	NMU	Number of secondary cosines	
LDAT(K+2)	XMU(I), I=1, NMU	Secondary cosines	
LDAT(K+2+NMU)	LMU(I), I=1, NMU	Location of data for each secondary cosine (relative to JXS(11) or JXS(19))	
Data for XMU(1) (let J=K+2+2*NMU):			
LDAT(J)	INTEP	Interpolation parameter between secondary energies (INTEP=1 is histogram, INTEP=2 is linear-linear)	
LDAT(J+1)	NPEP	Number of secondary energies	
LDAT(J+2)	EP(I), I=1, NPEP	Secondary energy grid	
LDAT(J+2+NPEP)	PDF(I), I=1, NPEP	Probability density function	
LDAT(J+2+2*NPEP)	CDF(I), I=1, NPEP	Cumulative density function	
Data for XMU(2)			
•			
•			
•			
Data for XMU(NMU)			
Data for EIN(2)			
•			
•			
•			

Data for EIN(NE)

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