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Title: NUCLEAR ACCIDENT DOSIMETRY STUDIES AT LOS ALAMOS NATIONAL LABORATORY

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Abstract - Two critical assemblies have been characterized at the Los Alamos Critical Experiments Facility (LACEF) for use in testing nuclear accident dosimeters and related devices. These device, Godiva IV and SHEBA II, have very different characteristics in both operation and emitted neutron energy spectra. The Godiva assembly is a bare metal fast burst device with a hard spectrum. This spectrum can be modified by use of several shields including steel, concrete, and plexiglas. The modified spectra vary in both average neutron energy and in the specific distribution of the neutron energies in the intermediate energy range. This makes for a very favorable test arrangement as the response ratios between different activation foils used in accident dosimeters are significantly altered such as the ratio between gold, copper, and sulfur elements. The SHEBA device is a solution assembly which has both a slow ramp and decay period and a much softer spectrum. The uncertainly introduced in the response of fast decay foils such as indium can therefore be evaluated into the test results. The neutron energy spectrum for each configuration was measured during low power operations with a multisphere system. These measurements were extended to high dose pulsed operation by use of TLDs, moderated TLDs, and special activation techniques.

The assemblies were used in the testing of several accident dosimetry devices in studies modeled after the Nuclear Accident Dosimetry Studies that were conducted at Oak Ridge National Laboratory for about 25 years using the Health Physics Research Reactor. It is our intention to

conduct these studies approximately annually for the evaluation of the nuclear accident dosimeter systems currently in use within the DOE, alternative systems used internationally, and new dosimeter designs being developed or considered for field application. Participation in selected studies will be open to all participants.

INTRODUCTION

The Los Alamos National Laboratory Critical Experiments Facility has a long history of experiments and studies related to nuclear accident analysis and criticality safety. Several of the devices that have been used in these studies include the Godiva series of bare metal assemblies, the Little Boy mock-up of the weapon used at Hiroshima, Big Ten and Flat Top reflected metal assemblies, and the SHEBA and SHEBA II solution assemblies.[1] Neutron spectra have been measured for these geometries and several others related to nuclear accidents such as the Y-12 accident at Oak Ridge, TN in 1959.[2,3] Unfortunately, much of the work was not made available through the open literature.

The Health Physics Research Reactor(HPRR), a bare metal assembly at Oak Ridge National Laboratory (ORNL); was also used for dosimetry, both routine and accident, for almost 25 years in addition to a large number of radiobiology, radiation damage to materials and electronics, and neutron transport experiments.[4] The assembly was used as a mock-up of both a nuclear weapon spectrum and a nuclear accident spectrum. A total of 22 Nuclear Accident Dosimeter Intercomparison Studies (NADs) were conducted at the HPRR in which nuclear accidents were simulated for the testing of nuclear accident dosimeters and methods.[5] The last NAD was conducted at ORNL in 1985. Generally the studies were conducted for the benefit of the U.S. Department of Energy(DOE) facilities but several were conducted jointly with the IAEA and other related facilities around the world. Most of the work done on HPRR has been well documented in the open literature.

Since use of the HPRR was discontinued in 1985, there have been very few opportunities for the training of nuclear accident dosimetrists and the testing of dosimetry related equipment for DOE facilities. In June, 1995 the Los Alamos National Laboratory (LANL), under sponsorship of the

Office of Worker Safety and Hazards Management in the U.S. DOE, hosted an intercomparison study for DOE facilities. The work leading up to this study and the results of the study will be described.

FACILITIES

A number of the critical assemblies which have been constructed and used for benchmarking critical geometries are still operational at the Critical Experiments Facility. Most of the devices are permanently mounted inside heavily shielded concrete buildings call Kivas. As such, it is very difficult to characterize the neutrons fields due to the large amount of backscattering from the concrete. Although this backscattered field can be easily quantified, the effect of the field on the response of the various instruments and dosimeters to be tested is more difficult to predict or measure. This question of scattered neutron radiation in both routine and accident dosimeter testing has been a point of controversy and concern. The philosophy adapted for this work was to minimize the scattered component in relation to the direct radiation as much as possible but to include the remaining scattered neutron spectrum in the reference spectrum. Although it has not yet been quantified, the scattered component is assumed to be dominated in the subject geometries by scattering near or behind the source as opposed to being dominated by scattered neutron radiation from behind the detector from the source. With these considerations in mind, it was decided to limit the devices currently used for dosimeter testing to the Godiva IV assembly and the Solution High Energy Burst Assembly (SHEBA II).

Godiva IV is a fast burst bare metal assembly made from approximately 66 kg of highly enriched uranium. A typical high level burst has a full-width at half-maximum of less than 50 μs and a leakage neutron spectrum close to that of a pure fission spectrum with a delivered absorbed dose to tissue of up to 2 Gy at 3 m during each burst. During dosimetry testing, the assembly is located in an open high-bay door approximately 3 m wide.(Figure 1) Thick concrete walls are located about 2 m away on each side of the device inside the doorway. Dosimeters are arranged on an arc outside the doorway at 3 m from the centerline of the assembly. Both the assembly and the dosimeters are about 2 m from the concrete floor. Scattering in this geometry should be dominated by scattering from the floor and the concrete walls with some higher energy scattering coming from the 1-2 cm of steel in the doors. Future plans are to operate the device outside the doorway far enough to reduce the wall and door scattering to a negligible position of the total fluence. Portable shields were used to modify the basic spectrum to simulate more varied accident spectra. Details of the shield design can be found in Reference 7.

SHEBA II is a solution assembly located in a relatively small, light sheet metal building.(Figure 2) SHEBA has a 49 cm inside diameter containment tank which goes critical with about 44 cm of low enriched uranyl fluoride.[8] Control of the assembly is maintained by adjusting the solution height during operation. For accident dosimetry testing, the device is operated either at delayed critical with a constant neutron leakage rate, on a controlled period to a predetermined delivered dose, or in free run mode where the solution is brought well above delayed critical and allowed to increase the reaction rate until the formation of electrolytic gases and increasing temperature causes the reactor period to increase rapidly through infinity and then quickly become negative. The later procedure provides a very realistic simulation of a probable process-line criticality accident. Typically the simulation develops slowly over five to ten minutes but then terminates rapidly. Although the operation could continue through the buildup into another peak as the gases clear and the temperature drops, the reaction is normally terminated at this point. Again the dosimeters are arranged on an arc at a distance of 3 m from the assembly centerline. The dominate scattering source is from the ground in this case, which contributes a small but nonnegligible component which can not be corrected in an intercomparison of this type. Indications of the magnitude of the scattered field are seen in the deviation of the field from $1/r^2$ dependence when measured at 3 m and 6 m.

Counting facilities were provided for the NAD participants although some brought their own equipment or shipped the samples back to their facilities. A number of HPGe detectors were used to analyze the activated foils. About half of the detectors were 30% and the rest 110% efficient. These were surrounded by lead bricks to minimize increased background due to the number of activated foils in the area. Sulfur pellets were processed by first reducing the pellet through burning on a hotplate and then counting the residue on an anthracene beta spectrometer. Beta proportional counters were also available. Electronic scales were provided with an appropriate range although some of the foils used were so small that an accurate determination of the mass was questionable in a working environment.

PARTICIPATION

A total of 17 representatives from 10 facilities participated in the NAD-23 Intercomparison Study. Participants ranged from the very inexperienced to individuals who had participated in previous NAD Studies and had many years of experience. Results were reported by all participants except one. Two of the reported results represented the combined efforts of two facilities in one case and three in the other. In both cases, the facilities used the same dosimeter designs, algorithms, and had cooperative agreements in dosimeter development or processing. All of the participants used activation foils except one, who relied on TLDs in combination with published accident spectra. The reported results do not indicate the facility, participant, or dosimeter type as this exercise was considered to be a learning workshop and not a test of the dosimeter capabilities or the participants' abilities. It is expected that a breakdown of the performance of the dosimeter types can be made after future NAD Studies when the available data is statistically more significant. Even then, it must be kept in mind that the participants' level of experience has a high impact on the reported results and therefore the resultant data should not be considered conclusive of the performance to be expected from a specific dosimeter design.

NEUTRON FIELD CHARACTERIZATION

The neutron field characterization was done by using a combination of detector systems and code calculations. The neutron spectrum was measured/calculated using the LANL Bonner Sphere Spectrometer System. These measurements were then used to refine neutron spectra that were calculated from Monte Carlo codes, measured at other facilities, or derived from other measurements in prior experiments. The normal unfolding technique applied to Bonner sphere data can be considered a perturbation technique in this case, as the starting spectrum input into the unfolding code was a reasonable approximation of the actual spectrum. The results were then used to calculate the absorbed dose. For the SHEBA derived spectrum, it was possible to check the absorbed dose results against tissue equivalent proportional counter measurements for consistency. Derived neutron spectra were also compared to previous measured spectra from the same assembly or ones very closely related, e.g., the HPRR published spectra.(Figure 3,4)

Several detectors were located in fixed positions such that the outputs could be used to normalize each of the many operations required, both for spectroscopy and for instrument response. The measured spectra were used to derive spectral responses for sulfur pellets and TLD based dosimeters. Two sulfur pellet types were used. A large size (~20 gm) was used to increase the sensitivity such that dose estimates could be made from less than 1 rad to well over 1000 rad. A smaller pellet (~5 cm), which provided more precise measurement within a limited range of about 10 to 100 rad, was used to make estimates of the delivered absorbed dose during some of the burst operations. TLDs were used in each case to provide the primary estimate of the absorbed dose. Estimates derived from the TLD data introduced the dominant error in the calculations because of increased variations in the supralinear region of the TLD response curve and due to the normal experimental variation inherent in these systems. Delivered doses derived for NAD-23 are shown in Table 1.

Reported results from accident dosimeters tested in each configuration was evaluated to insure that the reference values were consistent. In all cases, the reported results from several of the experienced participants in NAD-23 were in excellent agreement with the reference values although individual variations were sometimes considerable.

TESTING PROCEDURES

In general, accident dosimeters were mounted on 40x40x15 cm polymethyl metha acrylate (PMMA) phantoms. Care was taken to insure that the active elements of the dosimeters were no closer to the phantom edge than 10 cm and at least 2.5 cm from each other. In some cases an area dosimeter was used in conjunction with the personnel dosimeter. These were mounted without phantoms. Phantoms were placed at least 30 cm apart to minimize cross talk as much as practical. The 3 m distance from the centerline of the critical assembly to the front face of the phantom was the same as from the centerline to the center of the Bonner spheres during characterization. Care was taken to minimize complicating factors such as changes in surrounding areas which may cause changes to scattering. As a result, each experiment is considered to be reproducible with reasonable accuracy. Dosimeters were retrieved from the experimental area within about 30 minutes of the exposure. Generally, the foil activity measurements commenced about 1 hour following exposure.

RESULTS

The exercises were conducted in the order shown in Table 1. The first exercise using

SHEBA was intended to allow the participants to become familiar with the equipment and procedures as well as practice their analysis. In the second SHEBA run, the field was the same but the dose was increased substantially to allow for better counting statistics and a more realistic accident scenario. The neutron spectrum and the estimated neutron dose were provided to all participants for all the exercises except the final test with Godiva. The first Godiva test was conducted without shielding. This exercise provided the highest delivered absorbed dose in the study. One facility did not participate due to an administrative oversight. A similar test was conducted at a later date with that participant. This test provided a basic fission-like spectrum without significant moderation or scatter.

The second Godiva exercise involved use of a concrete shield which simulated the effect that may be expected from a wall or radiation shield. Following this test, the SHEBA device was again used, but this time in the "free run" mode of operation. The assembly was filled with a set critical amount of solution, as would occur in a real accident, and allowed to proceed through a complete cycle without operator intervention. The result is a long burst which does not have a set time mark from which to decay correct the results. The next exercise involved use of the Godiva assembly with Lucite shields. This highly thermalized spectrum is perhaps the most difficult to analyze for systems which use thermal neutron sensitive detectors, which included all of the NAD-23 participants. The intent of the test is to simulate the effect of water around the source.

The final exercise involved the Godiva assembly with a iron shield. This spectrum is unique in the distribution of intermediate energy neutrons. The spectrum somewhat resembles that of deuterium-moderated Cf. This exercise was conducted as a blind test. Neither the spectrum nor the absorbed dose was provided to the participants. The actual test was conducted the week following the scheduled study. Dosimeters were measured by LANL personnel and the results sent to the participants for evaluation. Several of the participants requested that the dosimeters be shipped to them at their facilities. Although the shield configuration was known, which would allow consulting the corresponding HPRR spectrum, no other indication of the spectrum characteristics or absorbed dose was provided. All of the results of the analysis, as provided by the participants, is given in Table 2.

CONCLUSIONS

Results of the NAD-23 Intercomparison Study indicates that it is unlikely that a facility which has not maintained an expertise in nuclear accident dosimetry would be capable of providing useful data in a short period of time in the event of a real accident. Even with highly trained personnel, the systems tested may be challenged to provide performance within the guidelines of ANSI N13.3[8] or DOE Order 5480.11[9]. During the NAD Study, data from blood sodium and hair sulfur was rarely used. This type of data may be necessary to obtain reasonable results. Using a fixed algorithm from a formal procedure limits the evaluation of the results. The dosimetrist should be knowledgeable enough to recognize problems in the results and make corrections as necessary in order to obtain improvements. For example, a typical accident dosimeter contains gold foils with and without cadmium, a copper foil inside cadmium, and a sulfur pellet. Many designs also incorporate an indium foil with and without cadmium. The indium can be analyzed for the ^{115m}In neutron reaction product which is created with a threshold of about 1.5 MeV. The counting of this reaction product is sometimes very difficult and often has a very large associated measurement error. Should the data from the sulfur n,p reaction with a threshold at about 2.5 MeV not be consistent with the In data, the option to ignore the In should be considered. This is a major alteration of the algorithm in most cases and can be difficult to implement in a short period under the conditions expected during an accident.

Data from the first 22 NAD Studies indicates a strong correlation between improved dosimeter performance and the number of times the facility participated in the Studies. A considerable effort has been invested at LANL into developing critical assembly data suitable for testing accident and routine dosimeters. Effort has also been directed to improving and documenting the accident dosimeter system in use at this facility. As a result, Los Alamos is able to provide this capability to the radiation protection community in the form of NAD Studies, routine and accident dosimeter testing, and on-site support of research and development projects.

REFERENCES

1. Paxton, H. C. *Thirty-Five Years at Pajarito Canyon Site*, Los Alamos Scientific Laboratory Report LA-7121-H, Rev. (1981).
2. International Atomic Energy Agency *Compendium of Neutron Spectra and Detector Responses for Radiation Protection Purposes*, IAEA Technical Report Series No. 318, Vienna (1990).
3. Hurst, G.S., Ritchie, R.H. , and Emerson, L.C. *Accidental Radiation Excursion at the Oak Ridge Y-12 Plant-III. Determination of Radiation Doses*, Health Physics **2**, 121-133 (1959).
4. Sims, C.S. *Collection and Analysis of Health Physics Research Reactor Operational and Use Data*, Oak Ridge National Laboratory Report ORNL-6145 (1985).
5. Sims, C.S. and Dickson, H.W. *Neutron Dosimetry Intercomparison Studies*, Radiat. Prot. Dosim. **10**(1-4), 331-340 (1985).
6. Sims, C.S. and Killough, G.G. *Reference Dosimetry for Various Health Physics Research Reactor Spectra*, Oak Ridge National Laboratory Report ORNL-TM-7748 (1981).
7. Malenfant, R.E., Forehand, H.M., and Koelling, J.J. *SHEBA: A Solution Critical Assembly*, ANS Transactions, **35**, 279-280 (1980).
8. American National Standards Institute, *Dosimetry for Criticality Accidents*, ANSI N13.3 (1969).
9. U.S. Department of Energy, Radiation Protection for Occupational Workers, DOE Order 5480.11 (1988).

Assembly	Shield Material	Shield Thickness(cm)	Delivered Neutron Dose(RAD)	Average Errors‡
SHEBA	None	N/A	11	16
SHEBA	None	N/A	108	19
Godiva	None	N/A	200	27
Godiva	Concrete	20	34	16
SHEBA	None	N/A	94	16
Godiva	Lucite	12	26	23
Godiva	Steel	13	59	41

Table 1 Configuration of critical assemblies used in the conduction of NAD-22 including the delivered doses.

Assembly	Lab A	Lab B	Lab C	Lab D	Lab E	Lab F	Lab G
SHEBA	12 (9)	15.7 (43)	10.4 (5)	11.35 (3)	13 (18)	11.9 (8)	14 (27)
SHEBA	114 (5)	107 (1)	92 (15)	133 (23)	162 (50)	83.1 (23)	127 (18)
Godiva	272 (36)		196 (2)	166 (17)	284 (42)	211 (5)	84 (58)
Godiva	32 (6)	32.9 (3)	31 (9)	47 (38)	39 (15)	47.5 (40)	35 (3)
SHEBA	104 (11)	126 (34)	102 (8)	106 (13)	130 (38)	91 (3)	101 (7)
Godiva	32 (23)	30 (15)	24 (8)	35 (35)	30 (15)	28.5 (10)	40 (54)
Godiva	48 (19)	49 (17)	119 (102)	34 (42)	52 (12)	77.6 (31)	22 (63)
Average Errors†	18	9*	27	31	27	22	39

Table 2 Summary of results for NAD-22 giving reported doses in RADS with error from reference shown parenthesis.

‡ Averages of the errors shown in parenthesis for all 7 reported participants for the given configuration
†Average errors for each participant of the results from the second SHEBA run and all Godiva runs.

* The first Godiva result was not available due to an operational error at the facility.

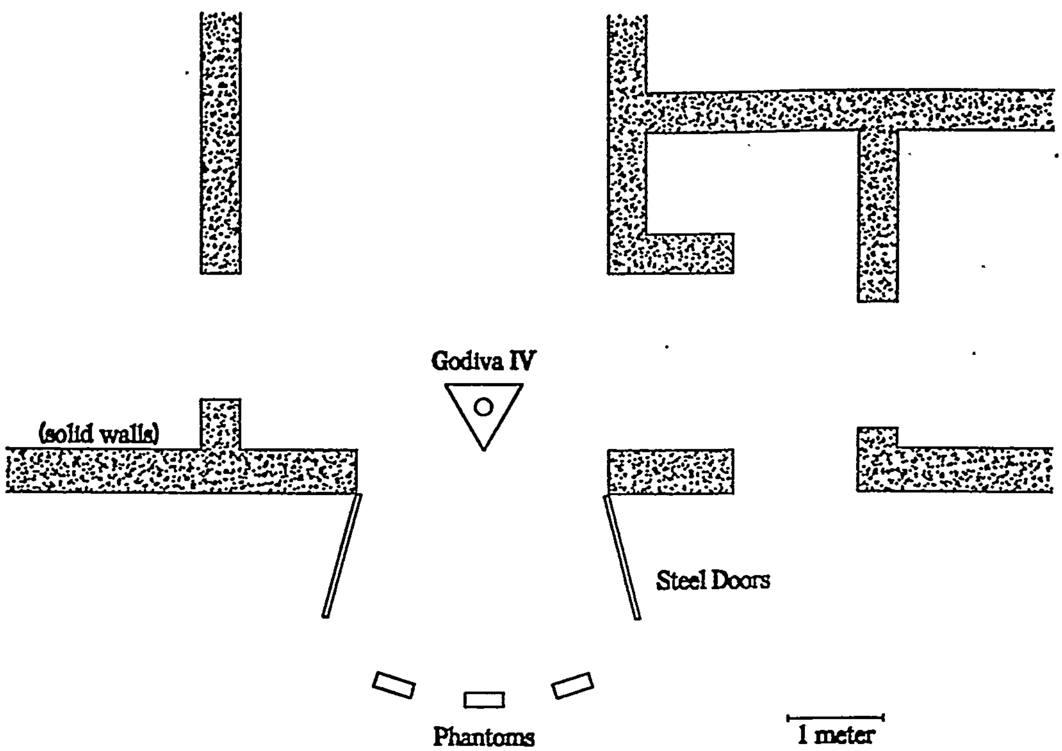

Figure 1 Floor plan showing location of Godiva inside building with phantoms located just outside during dosimetry testing.

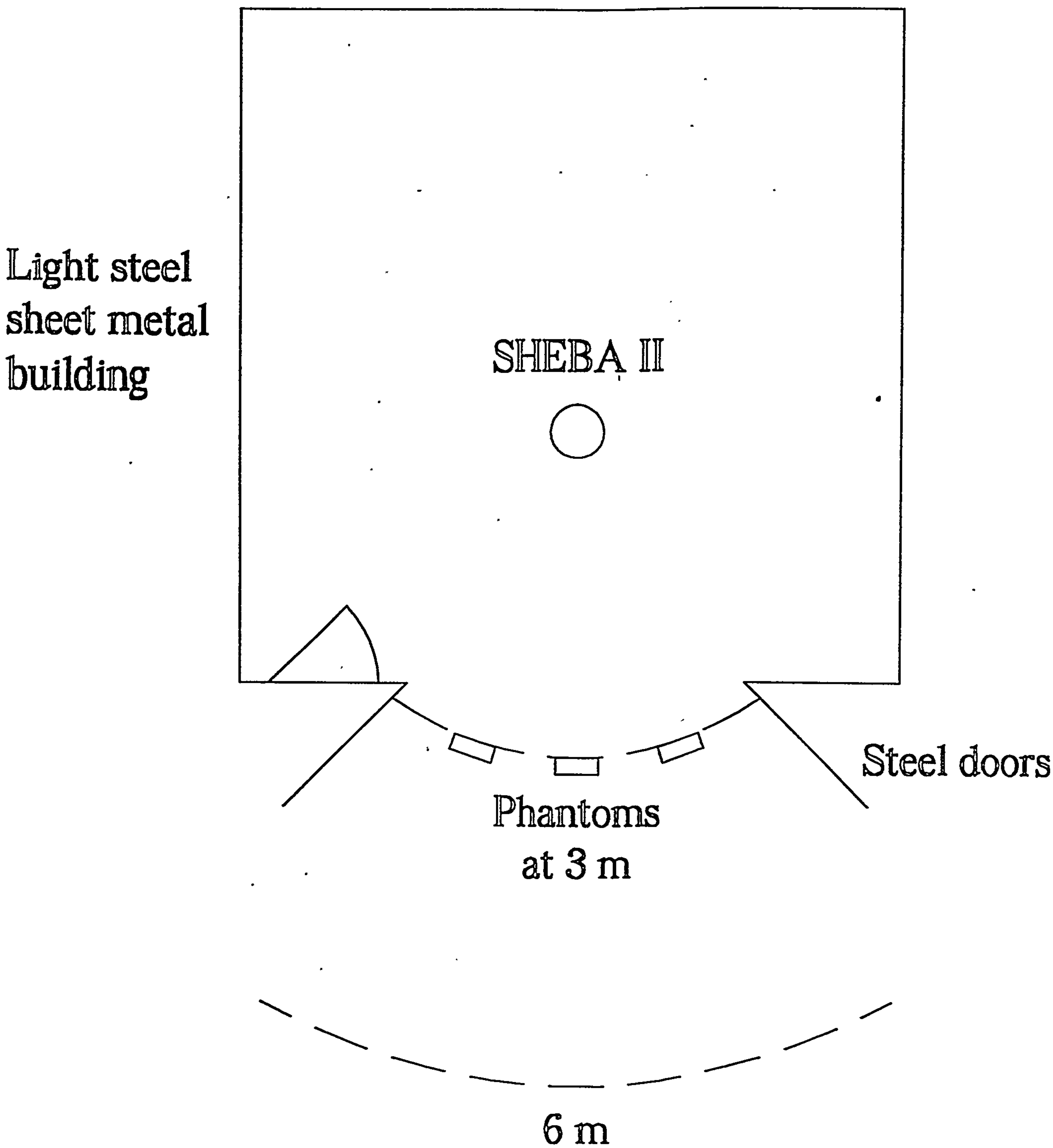

Figure 2 Floor plan showing location of phantoms with respect to SHEBA during dosimetry measurements.

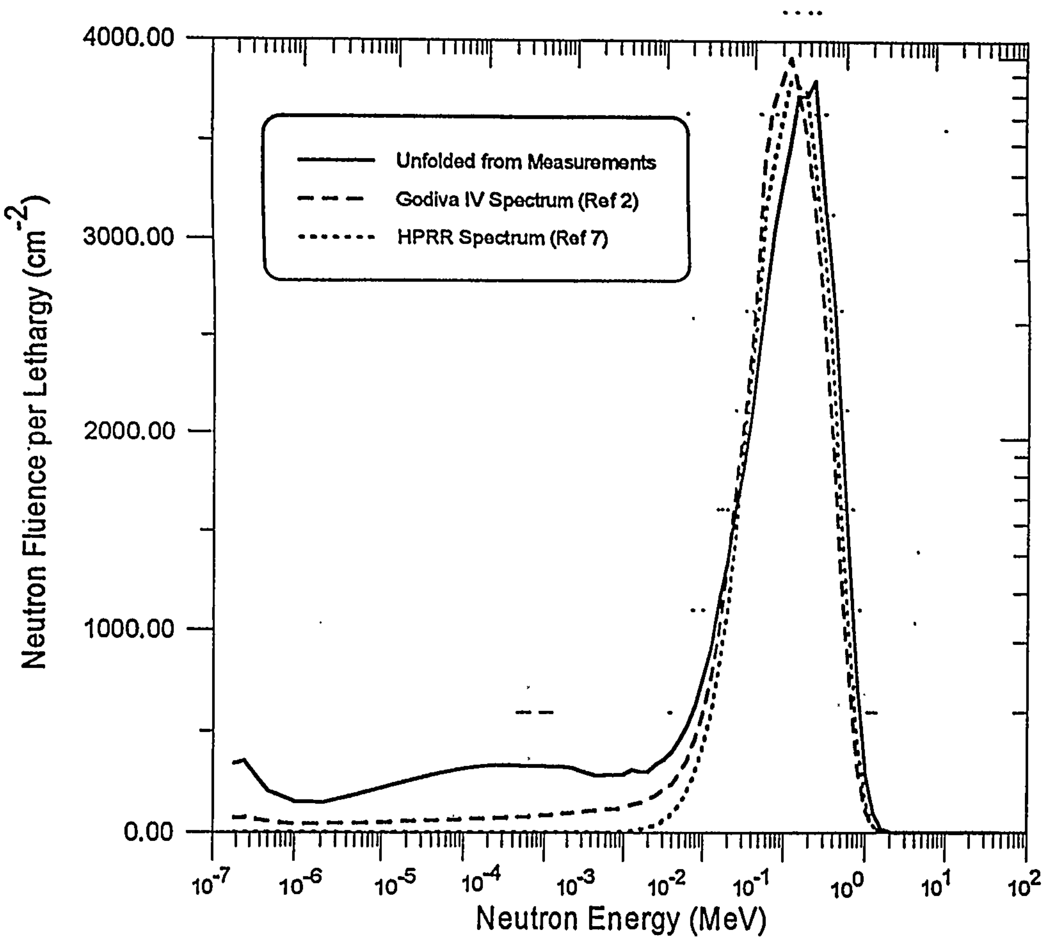

Figure 3 Neutron energy spectrum as derived by Bonner sphere measurements for Godiva without shielding. Comparison is made with two other published spectra.

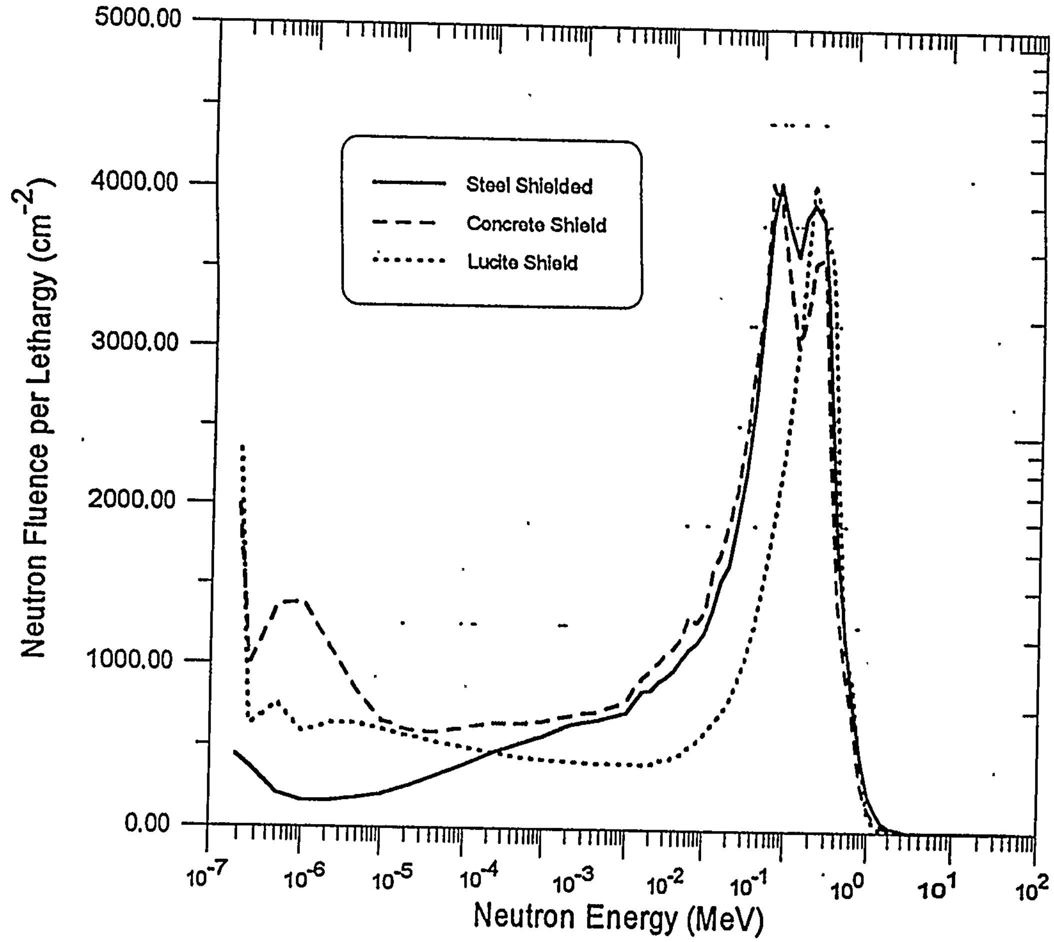

Figure 4 Neutron energy spectra as derived by Bonner sphere measurements for Godiva with shields used in dosimetry measurements.

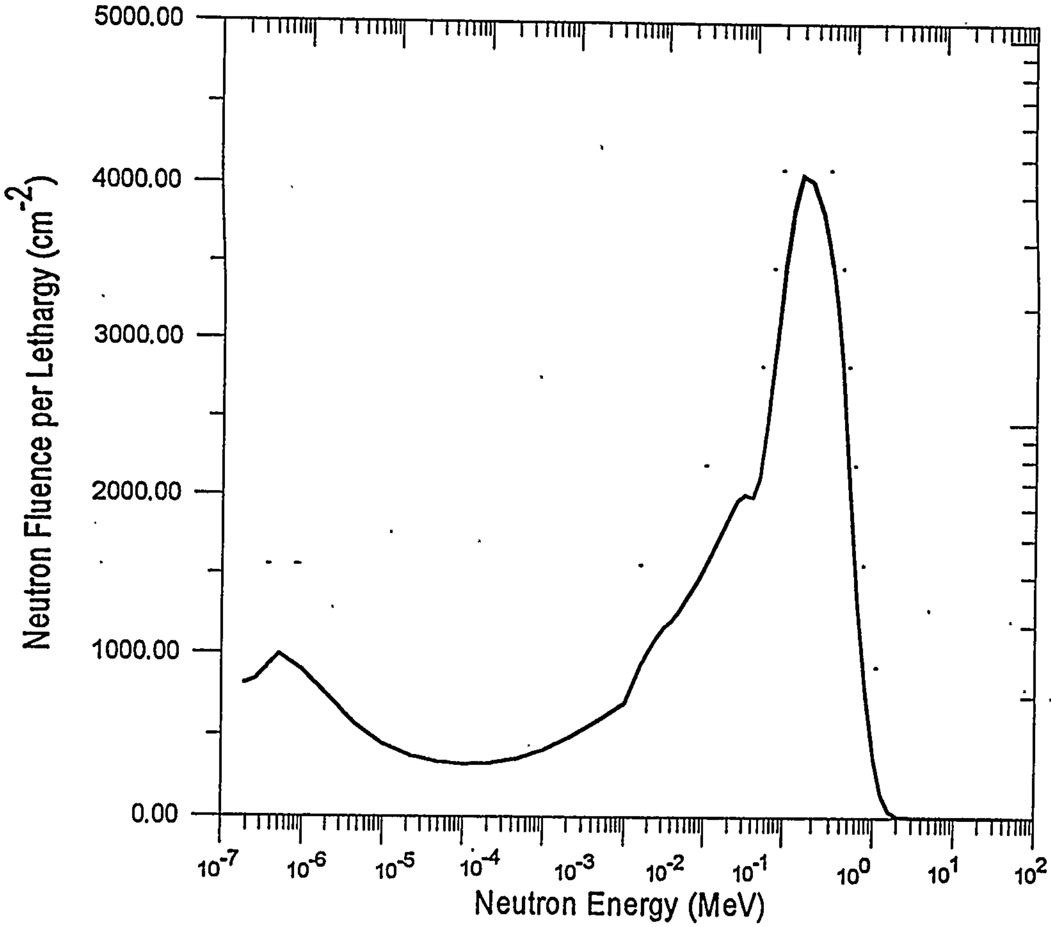

Figure 5 Neutron energy spectrum as derived by Bonner sphere measurements for SHEBA.