This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

MASTER
UCRL - 73550
PREPRINT

CONF-71125-3

RECEIVED BY DTIC DEC 7 1971

LAWRENCE LIVERMORE LABORATORY

UNIVERSITY OF CALIFORNIA
LIVERMORE

Livermore Pulsed Sphere Measurements Applied To Radiation Transport in Air

Luisa F. Hansen
John D. Anderson
John L. Kammerdiener
Calvin Wong

November 10, 1971

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

This paper was prepared for submission to
Radiation Shielding Information Center Seminar-Workshop
November 15-17, 1971

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

89

LIVERMORE PULSED SPHERE MEASUREMENTS APPLIED TO RADIATION TRANSPORT IN AIR *

Luisa F. Hansen, John D. Anderson, John L. Kammerdiener, & Calvin Wong
Lawrence Livermore Laboratory, Livermore, California 94550

Introduction

Since 1955 an appreciable and increasing effort has been invested in the study and calculations of the neutron and γ -radiation transport through air. The main emphasis has been on neutron transport, as evidenced by the number of such calculations found in the literature. However, many of the authors who have written neutron transport codes, such as French, Wells, Marcum, Straker, Plechaty and others, have also done γ -ray transport calculations. In the first volume of the Engineering Compendium on Radiation,¹ Clifford, Schaeffer and Clarke, and French and Wells have written review articles on photon scattering and neutron scattering in air, respectively. These papers discuss the different calculations done up to 1967 and describe the increasing complexity of the calculations as more realistic assumptions were taken for the interaction between the radiation and the medium. In a later report, Straker² extended these references up to 1970.

At Livermore (LLL), the air problem has a continuing interest since detailed information on the neutron and γ -ray transport is needed by different working groups. In the past the calculations were carried out mainly using the Monte Carlo transport code "SORS" written for neutrons³ and for photons.⁴ Since the beginning of this year these two codes have been replaced by two improved versions: TART⁵ for neutrons and TORTE⁶ for photons. A cross section evaluation group, headed by R. W. Howerton, is responsible for all the input cross sections used in these codes. In the LLL library (the "Howerton Library"⁷) one finds detailed information on all the cross sections used in any given calculation.

Our contribution to the neutron transport problem started at the end of 1967. The main impetus for our involvement was the disagreement found among the various neutron transport calculations. Fig. 1, taken
*Work performed under the auspices of the U.S. Atomic Energy Commission.

from an early report by Straker,⁸ shows this point very well. It was further felt that the correctness of any transport calculation could only be assessed by comparison with experiment. With this objective in mind, the Pulse Sphere Program was started and it was designed to meet the following requirements:

1. Measurements will be carried out for a simple geometry; the targets will be spheres of a single material or relatively simple compounds, to minimize the difficulties in the calculation due to a multi-dimensional media and complex materials.
2. The neutron spectra will be measured with relatively good energy resolution in order to be sensitive to the 14-MeV neutron cross sections. This was accomplished by using time-of-flight techniques, including bunching of the incident beam.

The $(d + t)$ reaction was chosen as the 14-MeV neutron source. Compared to the $(p + t)$ and the $(d + d)$ reactions, the $(d + t)$ reaction yields no break-up neutrons and is the closest to a mono-energetic and isotropic source. These features simplify the calculations problem and facilitate the interpretation of the measured neutron spectra.

To date, measurements have been carried out for a large number of materials from lithium to lead. Comparisons between calculations and measurements at Livermore and by other groups at Los Alamos and Oak Ridge, have shown the measurements to be a powerful check on the calculational methods. The adequacy of scattering model approximations due to lack of detailed information on inelastic processes, shape of angular distributions, and magnitude of cross sections, has been tested by comparing calculations with measurements. The problem of neutron transport in air was approached by making pulsed sphere measurements on liquid nitrogen and liquid oxygen as described in the next section.

Experimental Measurements and Calculations for Nitrogen and Oxygen

The nitrogen targets were spherical dewars of 10.5 cm, 19.05 cm, and 55.9 cm radius, equivalent to 0.58, 1.06, and 3.0 mean free paths respectively for 14-MeV neutrons. The oxygen measurements were made using the smallest dewar filled with liquid oxygen and corresponds to 0.72 m.f.p.

The measurements were divided into high energy measurements (from 1.6 MeV to 14.8 MeV) and low energy measurements (from 1 MeV down to 1 keV or less). The experimental techniques and apparatus have been described in detail elsewhere.^{9,10,11}

These experiments yielded emission spectra measurements for comparison with the predictions of the neutron transport calculated with SORS. Because of the serious disagreements found for both nitrogen and oxygen,^{9,10} steps were taken to improve the calculations. The major shortcoming was in the handling of inelastic scattering to discrete levels. Once these were included with their respective differential cross sections, the agreement between calculations and measurements improved greatly. Furthermore, revisions of the elastic and inelastic neutron cross sections to bring them in agreement with reported values found in the literature resulted in improved agreement between measurements and calculations.

Figs. 2-7 show a comparison between experimental measurements and SORS calculations. For nitrogen, Libraries¹⁰ A and B were considered because of the inconsistencies existing at that time with respect to the experimentally measured nitrogen cross sections. The measurements of the elastic and non-elastic cross sections by two different experimental groups did not add up to the higher precision measurements of the total cross section by a third group. Fig. 2 shows the predictions of Libraries A and B for the 0.58, 1.06, and 3.0 m.f.p. measurements while Fig. 3 shows Sauter and Robinson¹² calculations of the Gulf General Atomic (GGA)¹³ measurements for liquid nitrogen between 1 and 10 m.f.p., with these two libraries.

Early in 1970 Library C was chosen to replace A and B, because of the impracticality of having two libraries available to the users. In Library C the (n,n') cross sections to the continuum were decreased to improve the agreement with measurements below 3 MeV neutron energy. Shortly after Library C was formulated and tested against the measurements, Young and Foster¹⁴ re-evaluated the END/F library for nitrogen. Fig. 4 shows a comparison of SORS calculations carried out with Lib. C and Y-F library¹⁴ for the high energy measurements. Figs. 5 and 6 show a comparison between the calculations with Library C and the low energy measurements for the 3 m.f.p.

dewar and the GGA measurements¹³ between 1 and 10 m.f.p., respectively.

A detailed discussion and comparison of these four libraries (A, B, C, and Y-F) have been given extensively in the past^{10,15} and will not be repeated here. However, it is important to point out that the close agreement among the calculated results with the four libraries results from the fact that the elastic, inelastic, and total cross sections are practically the same for the high energy neutron groups ($13 < E_n < 14.8$ MeV). These calculations are not sensitive to differences of $< 30\%$ in the magnitude of the elastic and non-elastic cross sections between the various libraries for neutron energies between 7 and 11 MeV. The A and Y-F libraries have higher values for the elastic cross sections in the 7 to 11 MeV energy region, while the B and C libraries have higher non-elastic cross sections. Although the elastic and non-elastic cross sections differ, the total cross sections as a function of energy are the same in all libraries. Recently Perey and Kinney¹⁶ (1971) remeasured the neutron elastic scattering cross sections between 6 and 8.6 MeV and their results corroborate the higher values used in the A and Y-F libraries. These results resolve the long-standing controversy on the nitrogen cross sections in the intermediate energy range, and the Young-Foster evaluation¹⁴ can now be considered as the correct neutron library for nitrogen.

For oxygen much better agreement existed between the measurements of the partial and total cross sections, and it was therefore easier to formulate a single neutron library. Fig. 7 shows a comparison between SORS calculations and measurements for the low¹¹ and high¹⁰ energy emission spectra. The neutron cross sections, inelastic levels, angular distributions used for oxygen between 1.6 MeV and 14.8 MeV are given in Ref. 15.

To summarize the latest work, Fig. 8 shows a comparison between TART calculations carried out with the LLL libraries for nitrogen and oxygen and some of the measurements. (The LLL library for nitrogen is taken from Young and Foster's evaluations;¹⁴ for oxygen the cross sections are from Howerton's evaluation.⁷)

Neutron Transport in Air

The comparisons between the revised SORS calculations and the measurements shown in Figs. 2 and 7, show that the calculations have been

improved to the extent that they were able to reproduce quite well the neutron spectra from nitrogen bombarded with 14-MeV neutrons. The spectra cover the neutron range energy from 1 keV up to 15 MeV for 1 up to 10 m.f.p. of nitrogen. Because of the hazards associated with large amounts of liquid oxygen, measurements were made only for the smallest dewar, i.e. 0.72 m.f.p. oxygen. Fig. 7 shows the agreement obtained between calculations and measurements for 0.72 m.f.p. of oxygen.

The good agreement of O_2 and N_2 gave us confidence that the predictions for the neutron transport in air from a 14-MeV source will be quite reliable. The first calculations were carried out with Libraries¹⁰ A and B. Fig. 9 shows a comparison between the neutron doses calculated for an infinite air medium with constant density of 1.29 mg/cm^3 as predicted by SORS and Straker's earlier calculations.⁸ The much improved agreement was indeed gratifying. In contrast to the results shown in Fig. 1, the discrepancies this time, were no larger than 10 to 20% between the different calculations.

The neutron fluences, however, are more sensitive to the magnitude of the elastic and non-elastic neutron cross sections since fluence is a differential quantity, neutron/MeV, while flux and dose are related to an integral of the fluence over neutron energy. Differences of 30 to 50% were found between the fluences predicted with Libraries A and B. In the appendix of reference 9, these neutron fluences are tabulated for 1 through 10 m.f.p. of air and for different neutron energy groups. Fig. 10, taken from Sauter and Robinson,¹² shows the neutron fluences calculated for nitrogen using Libraries A and B. Library A gave consistently higher values for the fluences in the range of neutron energies where the magnitude of the elastic cross sections are higher than in Library B; above 12 MeV and below 3 MeV the two calculations were in close agreement.

A comparison between the neutron fluxes calculated with Library¹⁵ C and Straker and Grutzner's calculations¹⁷ for infinite air with a constant density of 1.11 mg/cm^3 , is shown in Fig. 11. (The neutron emission energy range shown is from 0.10 to 15.0 MeV.) Fig. 11 shows excellent agreement between the two calculations. Using TART and the cross sections from the LLL Library,⁷ Flechaty¹⁸ has recently repeated

these calculations and find equally good agreement with Straker and Grutzner's calculations.

The fluence calculations again exhibit a strong dependence upon the neutron cross sections used in the calculations. Fig. 12 shows a comparison of the fluences at 400 and 1500 meters calculated by Straker¹⁷ and SORS with Library C. At 400 meters the two calculations show discrepancies of factors of 2 between 3 and 13 MeV, with Straker's values being consistently higher. For the very fast neutrons (13 to 15 MeV), SORS-C predicts higher values by a factor of 4. At 1500 meters the two calculations are very similar to 10 MeV; for energies above 10 MeV SORS-C again predicts higher values for the fluence, as much as a factor of 5 at 14 MeV. However one must take into account the fact that these large factors of discrepancy at 14 MeV result mainly from the size of the energy intervals being used. If the last four high energy intervals in SORS are added to match the one used by Straker and Grutzner,⁷ one finds SORS results higher only by 4% at 400 m. and by a factor of 3.4 instead of 5 at 1500 m. These larger fluences for the high energy neutrons at 1500 meters have been corroborated by Plechaty's calculations with TART;¹⁸ for energies below 10 MeV TART calculations¹⁸ are in good agreement with Straker's calculations.¹⁷ From the comparison between measurements and calculations carried out earlier with SORS-C for nitrogen, it is felt that for the high energy neutrons SORS-C or TART are calculating the correct values. For energies below 10 MeV, SORS-C is underestimating the values of the fluence at closer distances since the calculated values are very close to those given by SORS-B,¹⁵ where the neutron elastic cross sections are known to be underestimated by as much as 30%.

Conclusions

From the comparison between the various calculations, it can be stated that the neutron transport in air from a 14-MeV neutron source is determined mainly by the neutron cross sections at energies close to the source energy. Integral quantities, such as flux and dose as a function of distance from the source, are insensitive to the values of the partial neutron cross sections below 12 MeV. This explains the good agreement

obtained with the different neutron libraries for the flux and dose since the partial and total neutron cross section for the high energy neutron group were very similar.

On the other hand the neutron fluences, as a function of distance to the source and for a given neutron energy group, are more sensitive to the cross sections. The calculations with Libraries A and B of the fluence in nitrogen differed by as much as 50%. In the region of 5 to 12 MeV, the fluences calculated by Library A were consistently higher by 20% over that calculated with B or C. This is a direct result of the higher elastic scattering cross sections used in Library A. For air the differences in the fluences calculated with A and B were of the order of 30%.

The above calculations were presented in Ref. 15, where it was also pointed out that the cross sections below 12 MeV were important in the calculations of the γ -ray transport in air. In this case, the intensity of the γ -ray flux is determined mainly by the higher or lower values taken for the inelastic cross sections. Perey and Kinney's recent measurements confirming the lower values for these cross sections, answer this question and validate the Young-Foster evaluation of the ENDF/B library.

Gamma ray fluences are more difficult to calculate, since they depend not only on the inelastic neutron cross sections, but also on the (n,p) , (n,d) , and (n,α) absorption cross sections. Measurements of the γ -ray fluence at 14 MeV and lower energies will determine the degree of hardening of the fluence as a result of the above cross sections.

References

1. C. E. Clifford (Pg. 246), N. M. Schaeffer (Pg. 253), E. T. Clarke (Pg. 255), and R. L. French and M. B. Wells (Pg. 350), Engineering Compendium on Radiation Shielding, Vol. I (Springer-Verlag, New York 1968).
2. E. A. Straker, Status of Neutron Transport in the Atmosphere, ORNL-TM-3065 (July 29, 1970).
3. E. F. Flechaty and J. Kimlinger, SORS Monte Carlo Neutron-Transport Code, UCRL-50532 (1968).
4. John Kimlinger, Ernest F. Flechaty, and John R. Terrall, SORS Monte Carlo Photon-Transport Code for the CDC 6600, UCRL-50358 (1967).
5. E. F. Flechaty and J. R. Kimlinger, TART Monte Carlo Neutron-Transport Code, UCIR-522 (February 1971).
6. E. F. Flechaty and J. R. Kimlinger, TORTE Monte Carlo Photon-Transport Code, UCIR-537 (June 1971).
7. R. Howerton, "Integrated System for Production of Neutronic and Photonic Calculated NAL Constants," UCRL-50400, Vol. IV (1970).
8. E. A. Straker, Calculations of the Transport of Neutrons from Fission and 14-MeV Point Sources in an Infinite Medium Air, ORNL-TM-1547(1966).
9. L. F. Hansen, J. D. Anderson, E. Goldberg, E. F. Flechaty, M. L. Stelts and C. Wong, "Time Spectra from Spheres Pulsed with 14-MeV Neutrons," Nucl. Sci. Eng., 35, 227-239 (1969).
10. L. F. Hansen, J. D. Anderson, E. Goldberg, J. Kammerdiener, E. Flechaty and C. Wong, "Predictions for Neutron Transport in Air, Based on Integral Measurements in Nitrogen and Oxygen at 14 MeV," Nucl. Sci. Eng. 40, 262-282 (1970).
11. P. Brown, J. D. Anderson, W. Webster and C. Wong, "Time Spectra of 1 eV to 1 MeV Neutrons from Spheres Pulsed with 14 MeV Neutrons," Proc. ANS Seventeenth Annual Mtg., 398 (Boston, 1970).
12. G. D. Sauter and P. A. Robinson, Calculations of the Transport of Neutrons from a (d,t) Source Through Mean Free Paths in Liquid Nitrogen, UCRL-50856 (1970).

13. J. L. Pigg and J. L. Russell, Jr., Neutron Penetration Measurements Program, AEC Report GA-8485 Gulf Atomic Inc. (1968).
J. L. Russell, Jr., Measurements and Analysis of the Transport of Fast Neutrons in Liquid Nitrogen, DASA Report GA-9081 Gulf Atomic Inc. (1968).
14. P. G. Young and D. G. Foster, Jr., "Nitrogen Evaluation ENDF-MAT-506," Los Alamos Scientific Laboratory (Private communication 1970).
15. L. F. Hansen, J. D. Anderson, J. L. Kammerdiener and C. Wong, Sensitivity of Monte Carlo Calculations to the Neutron Cross Sections for Neutron Transport in Nitrogen and in Air, UCRL-51031 (March 1971).
16. F. G. Perey and W. E. Kinney, "On a Possible Resolution of the Non-Elastic Cross Section Discrepancy Below 9 MeV," Oak Ridge National Laboratory (Private communication July 1971).
17. E. A. Straker and M. L. Grutzner, Neutron and Secondary γ -Rays Transport in Infinite Homogeneous Air, ORNL-4464 (1969).
18. E. F. Plechaty, TART Monte Carlo Calculations of 14.0 MeV Neutron Transport in Homogeneous Air, UCID-15849 (May 1971),

Figure Captions

Fig. 1 Comparison of the neutron doses versus range for a 14-MeV point source in an infinite medium of air of density 1.29 mg/cm^3 , as predicted by different calculations.

Fig. 2 Comparison between the measured neutron spectra from 0.58, 1.06, and 3.0 m.f.p. of liquid nitrogen and SORS calculations carried out with neutron Libraries A and B (Ref. 10).

Fig. 3 Comparisons of the detector-output pulse height spectra calculated using the neutron Libraries A and B, with the spectra from nitrogen obtained from the GGA measurements, for various source-detector distances. The numbers in parentheses are normalization factors. The GGA spectra shown have been normalized to the calculations with Library B (Ref. 12).

Fig. 4 Comparison between the measured neutron spectra from 0.58, 1.06, and 3.0 m.f.p. of liquid nitrogen and SORS calculations carried out with neutron libraries C and Young-Foster evaluation (Refs. 14 & 15).

Fig. 5 Comparison between the measured spectrum below 1-MeV neutron energy from 3.0 m.f.p. of liquid nitrogen and SORS calculations carried out with Library C (Ref. 14) for two nuclear temperatures of 1.25 and 1.54 MeV.

Fig. 6 Comparison between the GGA measurements of the neutron fluences in liquid nitrogen at distances of 50.8, 121.9, 152.4 and 183 cm from the 14-MeV source and SORS calculations carried out with Library C for nuclear temperatures of 0.98 and 1.54 MeV.

Fig. 7 Comparison between the measured neutron spectra below 1 MeV and above 1.6 MeV, from 0.72 m.f.p. of liquid oxygen and the predictions of the SORS calculations with the neutron library given in Ref. 15 for the low energy spectrum (See figure 6). The calculations were carried out for nuclear temperatures of 0.98 and 1.68 MeV.

Fig. 8 Comparison between the measured neutron spectra from 1.06 and 3.0 m.f.p. of liquid nitrogen and 0.72 m.f.p. of liquid oxygen and TART calculations carried out with the LLL Library.⁷ (The cross sections for nitrogen are from Young-Foster's evaluation¹⁴ and for oxygen from

Howerton's evaluation.⁷⁾

Fig. 9 Comparison of the neutron doses as a function of distance from a 14-MeV neutron source in an infinite air medium with constant density 1.29 mg/cm^3 , as predicted by SORS using Libraries A and B and by Straker's calculations.⁸

Fig. 10 Variation of neutron fluence with distance from an isotropic 14-MeV source in an infinite medium of liquid nitrogen, for neutrons in various energy groups. The curves are drawn only to show the trend of the data points; λ is a group attenuation length obtained from the slope of the curves (Ref. 12).

Fig. 11 Comparison of the neutron flux as function of distance from a 14-MeV source in an infinite air medium with constant density 1.11 mg/cm^3 as predicted by SORS using Library C (Ref. 15) and by Straker's calculations (Ref. 17).

Fig. 12 Comparison of the calculated fluences at 400 and 1500 m. from a 14-MeV source in an infinite air medium with constant density 11.1 mg/cm^3 as predicted by SORS using Library C (Ref. 15) and by Straker's calculations (Ref. 17).

Channel number

Channel number

NEUTRON TRANSPORT TO 400 METERS IN INFINITE HOMOGENEOUS AIR

NEUTRON TRANSPORT TO 1500 METERS IN INFINITE HOMOGENEOUS AIR