IS THERE STILL A NITROGEN CROSS-SECTION DISCREPANCY?

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ABSTRACT

For several years there have been great uncertainties as to which nitrogen neutron cross-section set should be used for neutron transport of 14-MeV neutrons in air. Various discrepancies were found between microscopic cross-section measurements and difficulties experienced in calculating correctly results of integral experiments. Most neutron air transport calculations have been performed using either the nitrogen cross-section set of Straker (1969) or the Young-Foster (1970) set. These two cross-section sets differ by about a factor of two in gamma-ray production cross sections around 14 MeV. In this short note we review the status of these evaluated cross-section sets on the basis of recent work and conclude that the Young-Foster evaluation, MAT 4133 Mod 2, is more consistent with all the available information and should be preferred over the Straker set.

In 1971 Straker¹ reviewed the adequacy of two different nitrogen evaluated cross-section sets for calculating several integral experiments. The two cross-section sets are (1) the Straker set² (1969) and (2) the Young-Foster set,³ the current ENDF/B MAT 4133 (1970). The integral experiments are (1) Operation HENRE⁴⁻⁷ (neutrons and gamma rays), (2) the GRT nitrogen Dewar experiments⁸⁻¹⁰ (neutrons and gamma rays) and (3) the LLL nitrogen Dewar experiment¹¹ (neutrons only). Although issued in August 1972, this report¹ summarizes work done at ORNL on the analyses of these experiments through the summer of 1971. The conclusions reached by Straker (considered still valid by most people active in this field today) were: (1) for neutron files, both sets agree reasonably well with all the data -- MAT 4133 appears to give better overall agreement and is, therefore, the preferred set; and (2) for the gamma-ray files, neither set fits the two gamma-ray experiments. Operation HENRE data are in good agreement with Straker's set but are underpredicted by MAT 4133 by 30 to 50%. However, Straker's set overpredicts the GRT experimental data by a factor of about 2, while MAT 4133 gives results about 40% too large.

Both the HENRE and particularly the GRT gamma-ray experiments are fairly sensitive to gamma rays from high-energy neutrons. For $E_n > 10$ MeV the two cross-section sets differ by slightly more than a factor of 2 in gamma-ray production cross sections. Since neither cross-section set fits both experiments, the choice between them has been difficult and somewhat arbitrary. MAT 4133 is a better compromise considering all the experiments, but if one wants not to underpredict gamma-ray fluxes, the Straker set would be preferred.

This situation has stimulated more work on gamma-ray production from nitrogen. Three new sets of data which have become available since Straker's conclusions were made are (1) a reanalysis with an improved unfolding code of the GRT nitrogen Dewar gamma-ray experimental spectra,¹² (2) the ORELA gamma-ray production data for nitrogen,¹³ and (3) data on a new spherical Dewar gamma-ray experiment at GRT.¹⁴ Although all of the data sets are not complete yet, it appears reasonable at this time to change the conclusions of Straker and strongly recommend the use of MAT 4133. It is the purpose of this note to give our reasons for doing so.

With reference to the first point, Reynolds and Sperling¹² have re-unfolded their spectra for the 1969 GRT gamma-ray Dewar experiment. In their 1969 data reduction, the results were presented for 11 discrete gamma-ray lines plus an essentially featureless continuum. The 1971 reanalysis was motivated by a desire to obtain more information concerning the high-energy part of this continuum and was, therefore, performed only for $E_\gamma > 4$ MeV. Their 1971 results for the line data are in very good agreement with their 1969 results, but for the continuum the new results are approximately twice as large as the 1969 data and show considerably more structure. With this increase in continuum data, their results are now in excellent agreement with the predictions from MAT 4133. To give an idea of the degree of quantitative agreement with the predictions of MAT 4133, using the figure for $D = 1.0$ foot in Ref. 12, we estimate $12.0 \pm 1.8 \times 10^{-6}$ photons/cm²/source neutron for the portion of the continuum in the range $4.0 \leq E_\gamma \leq 9.0$ MeV. To this we must add the flux from the line data (given in Ref. 12) which is $4.7 \pm 0.5 \times 10^{-6}$ photons/cm²/source neutron. Thus, the experimental yield for all gamma radiation having $E_\gamma > 4$ MeV, at 1.0 foot, is $16.7 \pm 1.9 \times 10^{-6}$ photons/cm²/source neutron. From the work of Straker and McGregor¹⁵ we

obtain, from their Fig. 4, for $E_\gamma > 4.0$ MeV a value of about 18.0×10^{-6} photons/cm²/source neutron at 1.0 foot for MAT 4133. Therefore, the notion that the GRT gamma-ray Dewar experiment indicates the presence of high-energy gamma rays not accounted for in MAT 4133 is not substantiated and is difficult to understand.

Several factors may have contributed to this notion. The initial transport calculation by Reynolds and Sperling,¹⁰ using the 1966 ENDF/B nitrogen neutron evaluation of C. W. Craven and the gamma-ray production data of Orphan et al.,¹⁶ failed to give these high-energy gamma rays. These authors concluded¹⁰ that their experiment indicated, at 14 MeV, an (n, α) cross section of over 200 mb, compared to 96 mb in MAT 4133, and that this difference in $\sigma(n,\alpha)$ would explain much of the nitrogen cross-section discrepancy we had previously observed.¹⁷ However, it is clear from the work of Straker and McGregor¹⁵ that collisions of neutrons with $E_n > 10$ MeV produce 91% of the total gamma-ray yield at 1 foot in the GRT Dewar experiment. Therefore, the nitrogen cross-section discrepancy observed¹⁷ for $E_n \leq 9$ MeV does not involve substantially increasing any (n, α) cross sections and is not in contradiction with the GRT Dewar results. The previous report¹⁸ on the resolution of this discrepancy in this energy range, therefore, is still valid.

As a second point, new ORELA nitrogen gamma-ray production measurements¹³ have been made and the results are in good agreement with MAT 4133, in particular, around $E_n = 14$ MeV. Furthermore, the structure observed in the 1971 unfolding of the 1969 GRT Dewar experiment is very similar to that observed in the ORELA data around $E_n = 14$ MeV and is correctly represented by MAT 4133. It would appear, therefore, that the 1971 unfolded results from the 1969 GRT

Dewar experiment are completely consistent with the ORELA nitrogen gamma-ray production measurement as well as ENDF/B MAT 4133.

In the fall of 1972, however, it appeared that the data on gamma-ray production from the new GRT spherical Dewar nitrogen sphere¹⁴ indicated much more gamma-ray production than predicted by MAT 4133 according to calculations performed independently at ORNL¹⁹ and at MAGI.²⁰ These indications were responsible for the revival of doubts that MAT 4133 had an adequate gamma-ray production file. However, it is now believed²¹ that the preliminary gamma-ray efficiencies originally suggested will be revised upward and that the "neutron-induced gamma-ray counts" in the detector are much larger than initially given.¹⁴ It appears now that these changes in efficiency and "neutron-induced gamma-ray counts" in the detector may be sufficient to bring the predictions using MAT 4133 into good agreement with the data. Since we do not yet have in hand all of the information needed to calculate this nitrogen sphere experiment, this conclusion can only be regarded as preliminary, although the indication still is that final data and the final calculations using MAT 4133 may not disagree.

We see from the above discussion that two of the three new data sets, namely the GRT nitrogen Dewar gamma-ray experiment reanalysis and the ORELA nitrogen gamma-ray production cross-section measurement, support the essential correctness of the gamma-ray file of MAT 4133. Analysis of the third experiment, the new GRT nitrogen sphere experiment, is not complete, but we can tentatively say that the data appear to support the correctness of the gamma-ray files in MAT 4133. The only known large failure of MAT 4133 is to correctly predict the gamma-ray doses from the HENRE experiment, the prediction being from 30 to 50% too small, whereas the Straker nitrogen cross-section set predicts correctly the HENRE gamma-ray data. However, as far

as is known, the only major difference between the Straker data set and MAT 4133 is for the gamma-ray production cross sections above $E_n = 10$ MeV. The Straker cross sections are approximately twice as large as those in MAT 4133, and they are inconsistent with the GRT nitrogen Dewar gamma-ray data and the ORELA nitrogen cross-section measurement. We must, therefore, conclude that the agreement between the predictions using the Straker cross-section set and the HENRE gamma-ray dose is fortuitous. The HENRE experiment is the only one which deals directly with neutron transport in the atmosphere and it is important to attempt to understand the nature of the disagreement seen with MAT 4133 for the gamma-ray dose. Because calculations using MAT 4133 agree with the other data mentioned above, it seems very unlikely that the failure of MAT 4133 in predicting the HENRE gamma-ray doses is related to gross errors (of the order of a factor of 2) in the gamma-ray production files, in particular at high energies. The use of the Straker gamma-ray file does not appear justified because of this one disagreement. More insight in the nature of this problem is currently under investigation¹⁹ using sensitivity analysis to ascertain if the differences observed between the calculated and observed gamma-ray dose in the HENRE experiment might be consistent with the cross-section uncertainties in MAT 4133.

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