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SCINTILLATION DETECTOR EFFICIENCIES FOR NEUTRONS IN THE ENERGY REGION ABOVE 20 MeV*

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ABSTRACT

The computer program SCINFUL (for SCINtillator FULl response) is a program designed to provide a calculated complete pulse-height response anticipated for neutrons being detected by either an NE-213 (liquid) scintillator or an NE-110 (solid) scintillator in the shape of a right circular cylinder. The point neutron source may be placed at any location with respect to the detector, even inside of it. The neutron source may be monoenergetic, or Maxwellian distributed, or distributed between chosen lower and upper bounds. The calculational method uses Monte Carlo techniques, and it is relativistically correct. Extensive comparisons with a variety of experimental data have been made. There is generally overall good agreement (less than 10% differences) of results from SCINFUL calculations with measured integral detector efficiencies for the design incident neutron energy range of 0.1 to 80 MeV. Calculations of differential detector responses, i.e., yield versus response pulse height, are generally within about 5% on the average for incident neutron energies between 16 and 50 MeV and for the upper 70% of the response pulse height. For incident neutron energies between 50 and 80 MeV, the calculated shape of the response agrees with measurements, but the calculations tend to underpredict the absolute values of the measured responses. Extension of the program to compute responses for incident neutron energies greater than 80 MeV will require new experimental data on neutron interactions with carbon.

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1. INTRODUCTION

The purpose of the computer program described in this report is to compute by Monte Carlo methods the complete response of an organic scintillation detector to incident neutrons. There are a number of existing computer programs [1-4] designed to predict, if not complete responses, at least the efficiencies of such detectors for a few selected thresholds. A well used, and often copied, program is the 20-year-old ORNL program O5S [1], a program well tested for incident neutron energies up to 20 MeV [3]. The program O5S was not designed for $E_n > 20$ MeV. Demanding a program to provide adequate computed responses for E_n up to 80 MeV required substantial new additions to the basic O5S programming as well as substantial revisions (for example, replacing non-relativistic by relativistic transformations). As a consequence, the present program is almost entirely new and warrants a new name: SCINFUL, for scintillator full response to neutron detection. This new program is divided into three, rather distinct, calculational phases. These are: (1) computing characteristics of energetic charged particles following neutron interactions with the carbon and hydrogen constituents of the detecting medium; (2) multiple scattering in and geometrical aspects of a finite-sized detector, including partial or complete absorption of the energy of the charged particles; and (3) the transformation of the total absorbed charged-particle energy into fluorescent light. A brief overview of each of these phases is given in the next sections, followed by comparisons of calculated efficiencies and total response spectra with experimental data. Lastly, measurements needed for program improvements and extension will be suggested.

2. COMPUTATIONS OF NUCLEAR INTERACTIONS

The basic methods used in the nuclear part of SCINFUL have been published [5]. Some comparisons with secondary-particle energy spectra obtained by Subramanian et al. [6] are shown in Fig. 1; a more complete set of comparisons with experimental data is given in a laboratory report [7]. The nuclear portion of SCINFUL is definitely semi-empirical. All available experimental data were utilized in developing the code, for example, published cross sections and angular distributions of reaction particles.* For some quantities, e.g., angular distributions, parametric representations of the data are devised and used for ease of interpolation and Monte-Carlo sampling. For cross sections not available from experiment, evaluated data from the U. S. ENDF/B evaluation [8] for $n + {}^{12}C$ reactions were used when available for $E_n \leq 20$ MeV. Other data, particularly for $E_n \geq 20$ MeV, were adapted from calculations using the statistical model code TNG [9]. Except for the

$$n + {}^{12}C \rightarrow n' + 3\alpha \tag{1}$$

reaction assigned as an "instantaneous" breakup of the ${}^{12}C$ into three α particles, the initial interaction is treated as a binary system:

$$n + {}^{12}C \rightarrow x + Y \tag{2}$$

where x represents a light reaction product, n, p, d, t, ${}^3He$, or α, and Y represents the heavy residual particle, Be, B, or C. SCINFUL computes deexcitation of a highly-excited Y particle using probabilities available from evaluation [10], experimental data where available, or statistical-model code calculations [9] when experimental data are not available.

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* Naturally occurring carbon has an isotopic composition of 98.9% in the isotope ${}^{12}C$ and 1.1% in the isotope ${}^{13}C$. The present version of SCINFUL approximates the carbon isotopic composition as being 100% of the ${}^{12}C$ isotope.

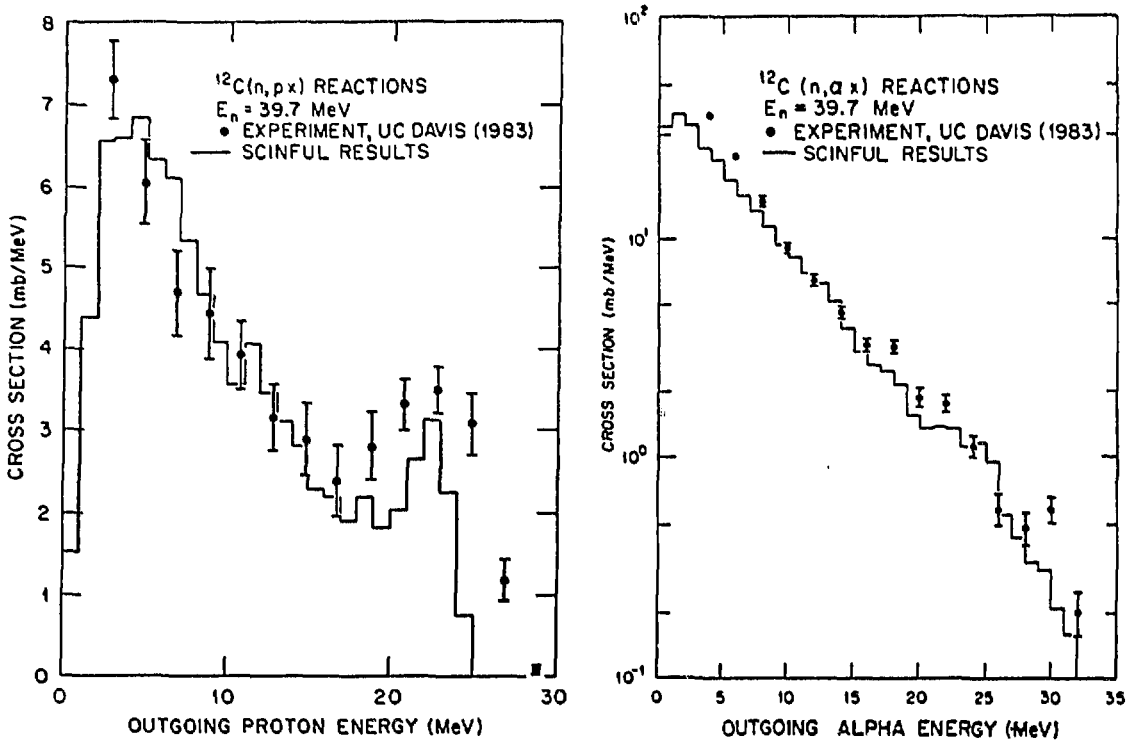

Fig. 1. Proton (left figure) and alpha (right figure) angle-integrated spectra for 39.7-MeV neutrons incident on ^{12}C as measured by Subramanian et al. (ref. 6) compared with calculations using techniques (ref. 5) in SCINFUL.

Secondary-particle energy spectra computed using SCINFUL have been extensively compared with theoretical spectra computed using an intra-nuclear cascade model by Brenner and Prael [11]. Comparisons were made for E_n between 15 and 60 MeV and these are exhibited in a recent laboratory report [12]. Two comparisons for $E_n = 40$ MeV are shown in Fig. 2. Although some differences are observed, particularly for low-energy secondary particles, it was concluded in Ref. 12 that the observed differences would have only minor effects on the total computed detector response.

3. MULTIPLE SCATTERING AND GEOMETRY EFFECTS

An organic scintillation detector combines an hydrogenous radiator with a scintillator into a single unit, such that a neutron interaction in the detector should result in fluorescent photons emanating from the detector to the photocathode of the photomultiplier tube. The output pulse from the photomultiplier tube, however, is not a monotonic function of the incident neutron's energy nor even of the amount of energy lost by the neutron in the detector (in the case that a scattered neutron subsequently escapes the detector). Indeed, for a series of separate interactions in the detector by a number of similar incident neutrons, the photomultiplier output pulses will range in amplitude from very nearly zero up to some maximal amount (which amount is monotonically, but not linearly, dependent upon incident neutron energy). The purpose of the program SCINFUL is to determine, as a computer "experiment," the expected pulse height distribution of these photomultiplier output pulses.

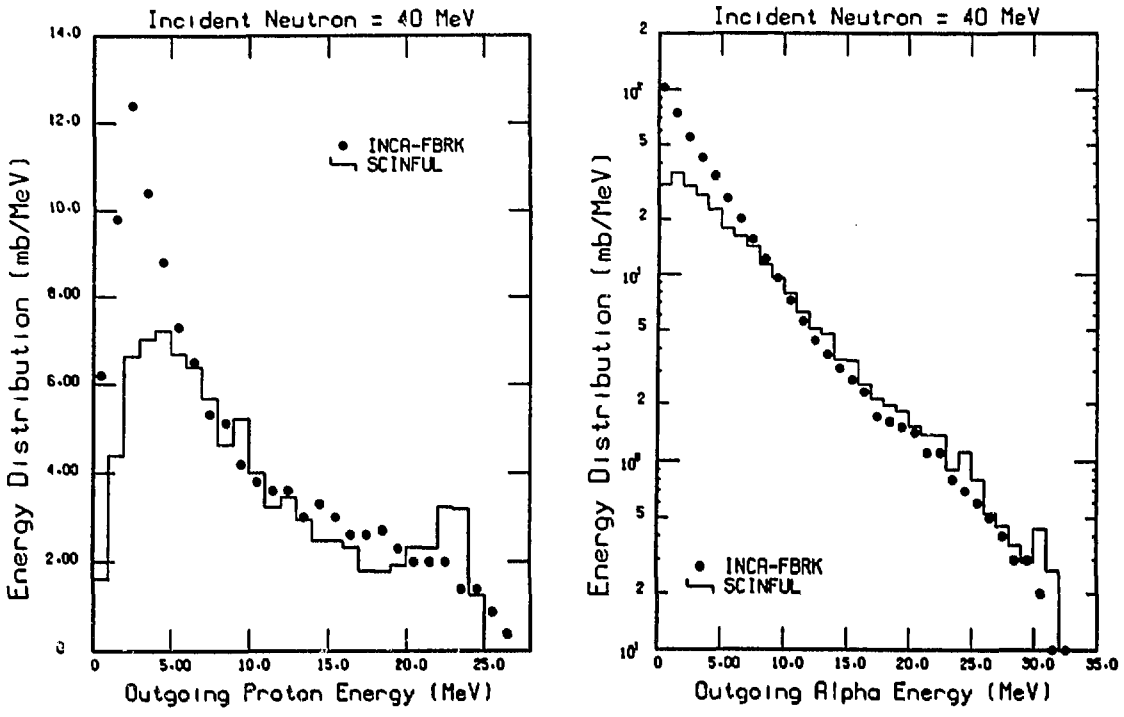

Fig. 2. Comparisons of outgoing secondary proton (left figure) and alpha (right figure) energy spectra for 40-MeV neutrons incident on ^{12}C. The solid points represent the spectrum computed using an intranuclear-cascade model code family INCA-FBRK (ref. 11). The histogram represents the same spectrum computed using techniques (ref. 5) in SCINFUL.

For detection of low-energy neutrons, the detector does not have to be very big to contain (i.e., slow down and stop) the recoil protons, and for a small detector most of the neutrons will scatter only once, or at most twice, in the detector. However, as the neutron energy extends above about 20 MeV, ranges of the recoil protons require larger detectors. Not only will multiple scattering of the incident neutron become more likely, but the relative probability of interaction with the carbon with respect to interaction with the hydrogen increases, and so modeling the various $n + {}^{12}C$ reactions (as discussed above) becomes important. As an example, consider a "history" for a high-energy incident neutron as schematically shown in Fig. 3. The neutron first has a collision with a hydrogen atom, and the scattered neutron traverses some path before colliding with a carbon atom. In the example, this latter collision is of the type ${}^{12}C(n,np){}^{11}B$. The secondary neutron is shown to interact once again, this time with a hydrogen atom, and then the neutron escapes the detector volume, and the "history" of collisions for this incident neutron is complete. Slowing down of each of the four charged particles results in photons being created by fluorescence, and some fraction of the total of the created photons will intercept the photocathode end of the detector.

In following the history exhibited in Fig. 3, one may observe several features that must be modeled in the program. The first task is to determine whether or not the first scattering takes place. If the Monte Carlo choice places an interaction inside of the detector, then the second task is to determine the type of interaction that took place, a task which requires cross-section data for all of the types of interactions energetically available.

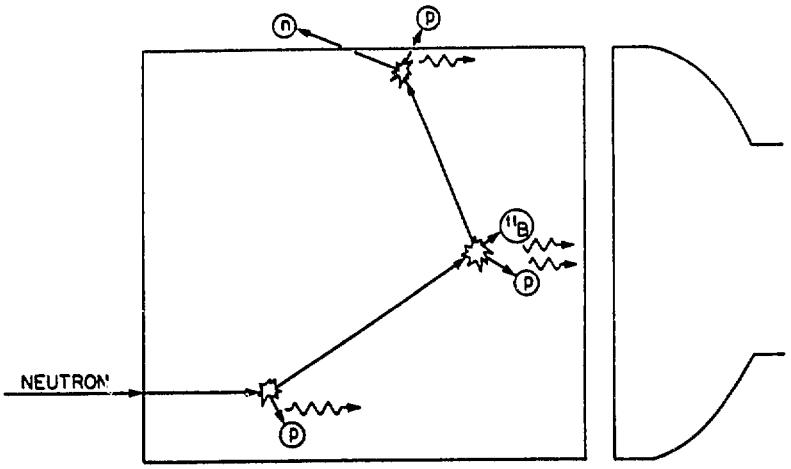

Fig. 3. Schematic illustration of a possible "history" for detection of a neutron in an organic scintillator. In this illustration the incoming neutron first collides with a hydrogen atom, and some of the kinetic energy of the subsequent recoil proton is converted into fluorescent light. The once-scattered neutron then strikes a ^{12}C atom, and the event postulated in this history is a $^{12}C(n,np)^{11}B$ reaction. Again, some of the kinetic energy of the recoiling ^{11}B ion and of the outgoing proton is converted into fluorescent light. The neutron leaving this reaction site collides with a hydrogen atom very near the surface of the detector. After this collision both the recoil proton and neutron escape the detector. Only a portion of the proton's initial kinetic energy is absorbed in the detector and so only that portion can interact with the organic molecules to produce fluorescent light. The escape of the neutron signals the end of the "history" of nuclear interactions by the neutron with the scintillation detector. All that remains is to determine the expected photomultiplier output pulse size by summing of the separate fluorescent yields.

The third task is to determine (again by Monte Carlo choice) the energetics and kinematics of the chosen reaction, and once having completed the reaction, to compute (a) the light output for each charged ion and (b) the direction and energy of the scattered neutron (if there is one) in the detector coordinates. Now the process just described is repeated for the scattered neutron, namely (a) determine a new probability for interaction, (b) determine a new path length to the surface of the detector along the scattered path, (c) determine by Monte Carlo if an interaction takes place inside the detector and, if so, what type, (d) do the energetics, kinematics, and fluorescent photon production by the charged ions, and (e) determine the second-scattered (in this example) neutron's energy and direction in the detector.

There are two other features exhibited in Fig. 3 that should be noted. The third scattering shows the recoil proton escaping the detector before all of its energy is lost. For high-energy neutrons, escapes of recoil protons are important, and so determining energetics of these processes is modeled in SCINFUL. Another feature shown in this figure is the observation that centers of fluorescent photon production may be at various places in the detector. Fluorescent photon absorption by the scintillator medium may be important, and so SCINFUL includes the capability of attenuating the light produced at the interaction point before it reaches the photocathode.

4. FLUORESCENT LIGHT CONSIDERATIONS

The third part of the neutron detection process is the absorption of the energy of the charged ion and the concomitant creation of light by excitation of the molecules of the scintillator. Although experimental work was performed in the early years of scintillation spectrometry to determine the fundamental physics of this phenomenon [13], the fact is that it remains the least understood part of the neutron detection process. Indeed, for modern users of scintillation detectors the important aspect is not the absolute quantification of fluorescent light that is created (in lumens, for example) but rather the quantification of the electronic output pulse of the photomultiplier tube. And although the quantification of the electronic output pulse can be made on an absolute basis in some electronic unit (say, volts), that unit is by itself not very useful since it depends upon the specific hardware of the measurement system. What is desired is a nuclear physics unit related to energy absorption by the detector which should be independent of the specific hardware; what has evolved is a hybrid labelled a "light" unit (L.U.), which is not light (i.e., photons) and which does depend, on an absolute scale, upon the material that makes up the scintillator as well as the characteristics of the photomultiplier tube. In an early definition, one "light" unit was the (essentially maximum) pulse height observed following detection of ^{60}Co gamma rays by the detector. More modern usage defines the "light" unit in terms of the electronic pulse height that would be observed upon detection of a monoenergetic 1-MeV electron by the scintillator. For detector calibration purposes, however, one usually uses gamma-ray sources for calibrations and takes the forward-scattered Compton electrons to be responsible for the largest electronic pulse heights observed from the photomultiplier tube.

One difficulty with the above-discussed calibration method is that it hides one aspect of the total neutron detection process, an aspect mentioned in passing during the discussion of Fig. 3, and that aspect has to do with the efficiency by which the system converts the energy of the fluorescent photons into electrons at the photocathode of the photomultiplier tube. A few experiments have been reported [14-17] which show that the efficiency for events which take place in the detector some distance from the photocathode is less than the efficiency for events which take place in the detector close to the photocathode. The usual calibration procedure results in a spread in sizes of output pulses which is then interpreted as a "resolution" effect due, for example, to fluctuations in the number of photoelectrons created at the photocathode following absorption of a specific amount of light. The loss of efficiency, on the other hand, relates to such aspects as the reflectivity at the surfaces of the scintillator. Fluorescent light attenuation affects the overall neutron-detection measurement differently [14] than do fluctuation effects. Folding in fluctuation effects, or "resolution" effects, results in modifying the "ideal" response in the region of the maximum-sized pulses. There will be pulses observed to be larger than one would observe in an "ideal" measurement; however, these will be compensated for by an equal loss of lower-energy pulses.

However, light attenuation always shifts the measured pulse height to smaller values; there can not be any pulses larger than the maximum obtained from an "ideal" measurement. Furthermore, the overall shape of the total measured response is different. One cannot measure the "ideal" response with just fluctuation effects folded in, and so one does not know the value of the maximum pulse height for the ideal case.

Now the point to this discussion may be stated, and that is that the many experimental reports [2,18-23] of "light" units versus proton (or other charged ion) energy are essentially devoid of any consideration of this problem of the efficiency of fluorescent light collection. In these experiments the detector is "calibrated" using gamma-ray sources [20], and then the pulse height with respect to said calibration is reported for different energy protons (neutrons) incident onto the face of the detector. The assumption is implicitly made that the incident protons (neutrons) will create light throughout the volume of the detector with

exactly the same probability as the calibrating gamma-ray sources create light throughout the volume of the detector, an assumption that is very likely to be wrong for most of the "light-unit" calibration measurements so far reported, and quite wrong for at least a few of them. It is little wonder, then, that there is poor agreement on the value of one "light" unit in the literature to within the usual assigned uncertainties.

These concerns bear upon the development of the light-unit conversions in SCINFUL. Light-unit values in NE-213 are tabulated [24] in SCINFUL for protons, deuterons, alphas, and carbon ions having energies between 0 and 100 MeV. The most important are for protons, and for these SCINFUL uses the values of light given by Verbinski et al. [25] for $0.1 \leq E_p < 40$ MeV; for E_p between 40 and 100 MeV the light-unit values were obtained from the current version of the O5S program [1] which are not reported in the Verbinski et al. paper [25]. Uwamino et al. [2] have reported measurements of proton light units $LU(p)$ for monoenergetic neutrons of 22.6, 27.6, and 48.7 MeV (no ΔE_n quoted). Their results, given in graphical form, are compared to $LU(p)$ used in SCINFUL in Table I.

Table I. Comparison of proton light-unit values used in SCINFUL with measurements of Uwamino et al. (ref. 2)

Proton energy (MeV)	$LU(p)$ SCINFUL	$LU(p)$ Measurement	Ratio S/M
22.7	12.61	12.08 ± 0.36	1.044
27.6	16.00	15.87 ± 0.48	1.008
48.7	30.99	31.15 ± 0.94	0.995

Light-unit values in SCINFUL for alpha particles also used the reported Verbinski et al. [25] and from the O5S program [1] values for E_α between 0.1 and 100 MeV. However, comparisons of computed responses with measured responses for E_n between 14 and 22 MeV indicated that for $E_\alpha \lesssim 10$ MeV, the Verbinski et al. [25] alpha-particle light-unit values were too small. The SCINFUL values are, therefore, somewhat larger so as to provide a computed detector response in better agreement with measurements.

Other charged-particle light-unit values in SCINFUL were estimated from measurements of Bechetti et al. [26] The values in SCINFUL were checked by comparing measured responses with computed responses. As best as can thus be determined the comparisons are in satisfactory agreement.

All of these tabulations are for the scintillator NE-213. For NE-110, Renner et al. [27] showed that proton light for this scintillator is the same as proton light for NE-213,* and carbon light is larger by about a factor of 3. In addition, comparisons of SCINFUL responses calculated for NE-110 with measured responses for NE-110 indicate that alpha light for NE-110 is about 75% larger than alpha light for NE-213, and that deuteron light for NE-110 is about 50% larger than deuteron light for NE-213. These increases are programmed into SCINFUL.

5. COMPARISONS WITH INTEGRAL DATA

Often, a neutron measurement will utilize a detector in an "integral" mode by setting a suitable discriminator (or bias) level on the electronic output of the phototube amplification

* This equivalence is an artifact of the definition of a "light" unit. According to the manufacturer (Nuclear Enterprises, Inc.) the absolute light output for NE-110 is ~75% of the absolute light output for NE-213.

system and scaling (counting) all registered pulse heights ≥ discriminator level. SCINFUL reproduces very well most reported integral measurements for $E_n < 20$ MeV. For $E_n > 20$ MeV, comparisons of SCINFUL calculations and two sets of measurements [28,29] using medium-volume detectors are exhibited in Fig. 4. The comparisons are very favorable. Note, however, that even for a low (and easily determined) bias of ~1 L.U. less than 50% of the total integral yield is used.

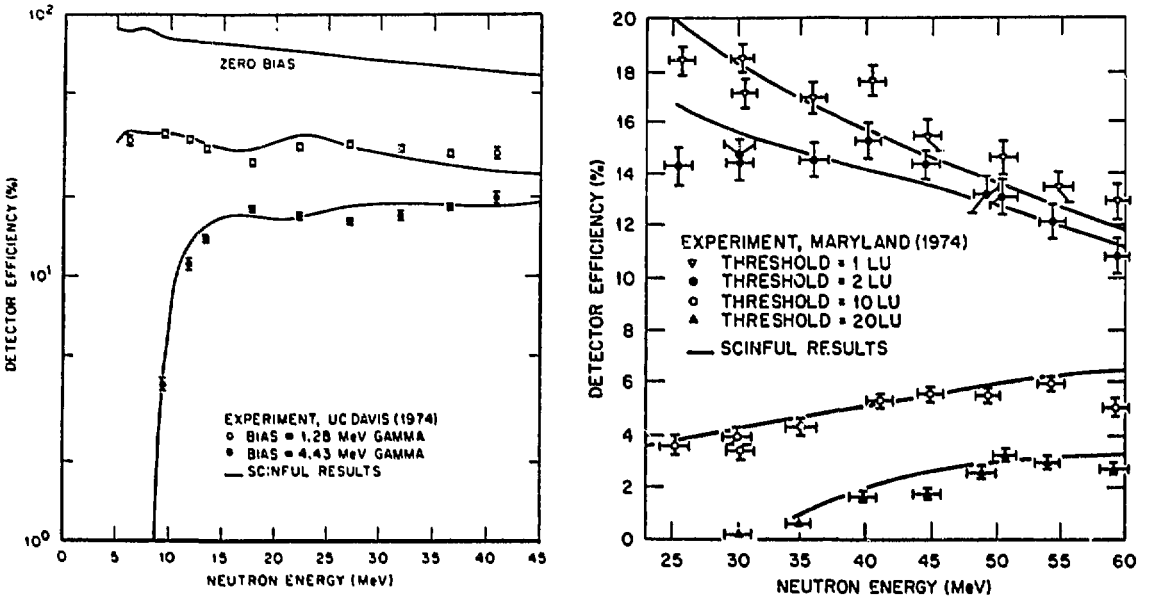

Fig. 4. Detector efficiency for an NE-102 scintillator as a function of neutron energy for the data of McNaughton et al. (ref. 28) for two different electronic thresholds (left figure) and for the data of Riddle et al. (ref. 29) for four different electronic thresholds (right figure). The SCINFUL calculations (ref. 7) are for NE-110, a scintillator very similar to NE-102. The detector of McNaughton et al. was 7.1 cm diam by 15.2 cm deep; the detector of Riddle et al. was 17.7 cm diam by 7.63 cm deep, and the incident beam was collimated to 5.1 cm diameter. This collimation was modeled in the SCINFUL program.

Table II. Efficiency comparisons with NE-213 measurements at Karlsruhe (ref. 30). Efficiencies are in percent.

E_n (MeV)	Experiment	SCINFUL	Ratio
	Bias = 0.6 MeV_e		
49.5	4.98 ± 0.15	4.71 ± 0.11	1.06 ± 0.04
75.4	3.82 ± 0.25	3.34 ± 0.08	1.14 ± 0.08
	Bias = 4.2 MeV_e		
49.5	4.01 ± 0.15	3.35 ± 0.09	1.19 ± 0.03
75.4	2.86 ± 0.25	2.71 ± 0.07	1.06 ± 0.10
	Bias = 17.5 MeV_e		
49.5	0.85 ± 0.10	0.78 ± 0.05	1.10 ± 0.15
75.4	1.06 ± 0.10	0.95 ± 0.04	1.12 ± 0.12

Similar measurements using a "thin" detector have the advantage of lessening multiple-scattering effects but have the disadvantage of charged-particle escape. Measurements made at Karlsruhe [30] using a 3-cm-thick NE-213 detector (thus having a large proton-escape contribution) are compared with SCINFUL calculations in Table II. The comparisons are favorable for all three bias levels at both neutron energies.

6. COMPARISONS WITH SPECTRAL DATA

Very few experimental spectral data exist in the literature for $E_n > 20$ MeV. Two available spectra [29] are exhibited in Fig. 5 compared with results of SCINFUL calculations. The shapes of the largest pulse height portions of both spectra suggested including a small amount of fluorescent light attenuation in the calculation, and so calculations were performed using an exponential attenuation having a small coefficient of 0.01/cm. Then the gains, in pulse-height light-units vs channel number, were adjusted at the largest pulse heights in order to display the calculated responses with the experimental responses. The agreement for $E_n = 30$ MeV in Fig. 5 is very satisfying. The comparison for $E_n = 45$ MeV in this figure is not so favorable, particularly for channels 60 through 80.

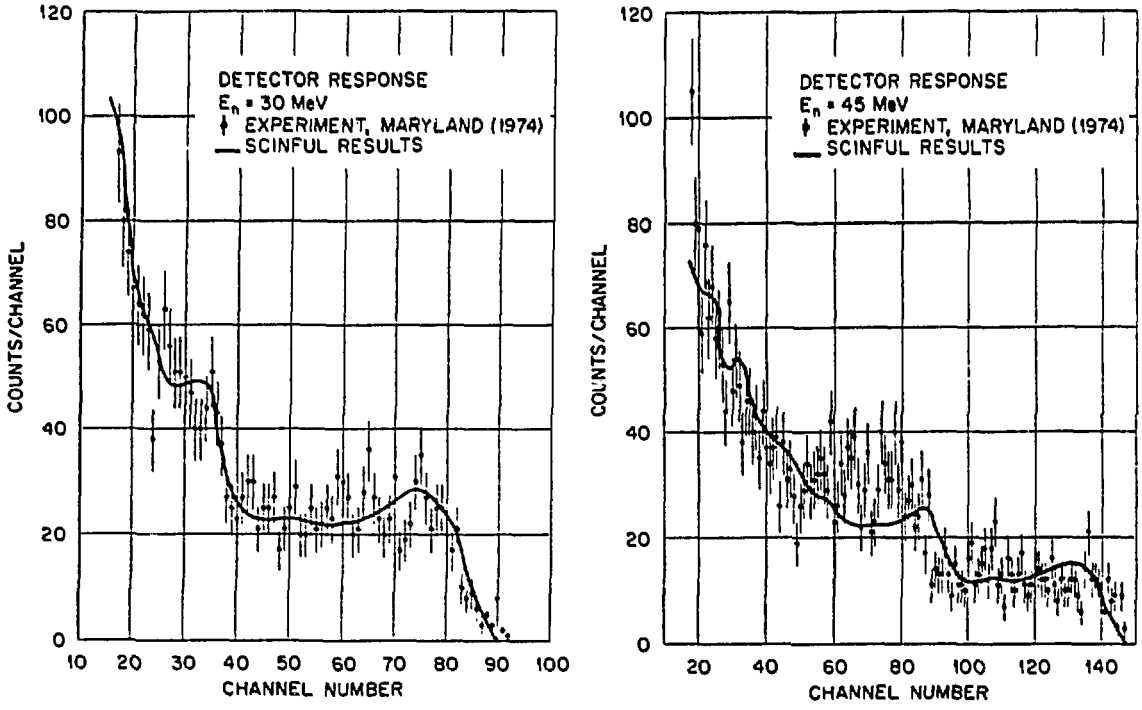

Fig. 5. Comparisons of experimental response measurements at $E_n = 30$ MeV (left figure) and 45 MeV (right figure) of Riddle et al. (ref. 29) and the results of SCINFUL calculations (ref. 7). Detector and neutron beam dimensions were as given in the caption to Fig. 4.

The other response experiment to be discussed is that of Lockwood et al. [31] at Michigan State. These experimenters used pulse-shape-discrimination electronic techniques not only to eliminate gamma-ray interactions, but also to attempt to electronically eliminate alpha interactions, leaving only the proton interactions in the detector to be recorded. Their hope was in this manner for E_n between 25 and 75 MeV to isolate the contributions

to the response from the protons created in the $n + {}^{12}C \rightarrow p + \ldots$ reactions and thus determine cross sections for this reaction channel.

There were some difficulties in comparing SCINFUL results with these experimental responses. The major one was the "calibration" of the pulse-height axis, for it appeared that the experiment was yielding protons of energies larger than the available energy. Furthermore, it was going to be difficult to determine a resolution function that might be used to fold resolution effects into the SCINFUL results for a better comparison with the measurements. Ultimately it was decided (1) to revise the calibration of the experimental proton-energy axis by about 8% and (2) to forego folding in of a response resolution function.

The initial SCINFUL calculations for comparisons with these data were set to predict only light output from proton events but it soon became evident that it was necessary to include all $Z = 1$ ions to obtain results of magnitudes similar to measurements. Second-round SCINFUL results for $E_n = 39.4$ and 74.3 MeV are shown in Fig. 6. The comparison for $E_n = 39.4$ MeV is quite favorable both in shape and absolute amplitude. For $E_n = 74.3$ MeV, the overall shape of the experiment is well reproduced by the calculation but the latter is ~20% smaller in magnitude than the former.

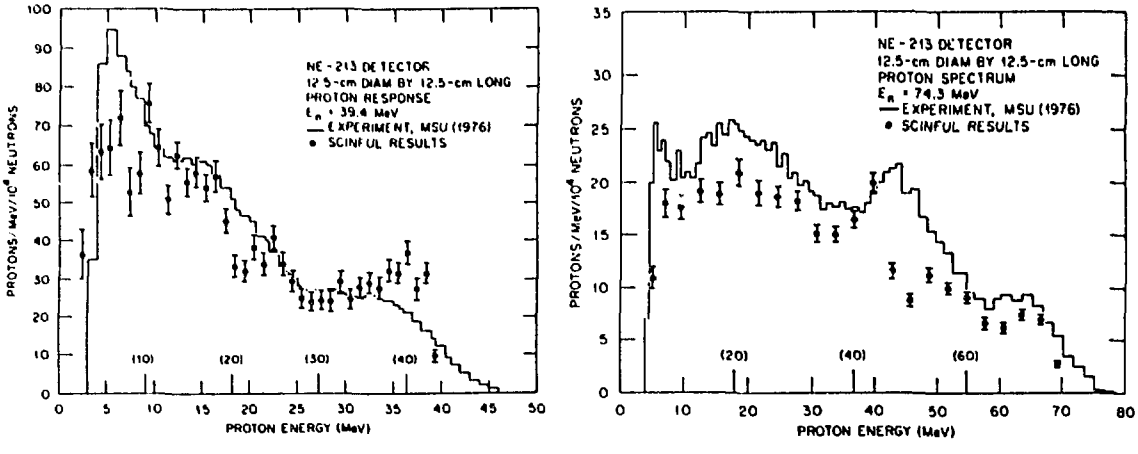

Fig. 6. Comparisons of experimental response measurements for $E_n = 39.4$ MeV (left figure) and 74.3 MeV (right figure) by Lockwood et al. (ref. 31) and results of SCINFUL calculations (ref. 7). The experimental pulse-height calibration for these data appears to be in error by about 8%. The small figures in parentheses indicate the pulse-height calibration given in ref. 31.

7. CONCLUSIONS AND RECOMMENDATIONS

The overall accuracy of SCINFUL calculations on an absolute scale depends on accuracies associated with evaluated data for $n+p$ scattering and for $n+{}^{12}C$ total cross sections. Indeed, for $E_n > 40$ MeV, the latter is the more important not only because of the increase in secondary protons from $n+{}^{12}C$ reactions with respect to $n+p$ scattering, but also, as may be observed for the $E_n = 74.3$ MeV comparison, the definitely observed contribution from the $n+p$ scattering represents <15% of the total pulse height spectrum (i.e., for

$E_p > 60$ MeV in the figure). Furthermore, it becomes more difficult with increasing E_n to delineate the $n + p$ contribution from the main portion of the total response.

Accurate measurements of $n+^{12}C$ cross sections are needed. In addition, measurements of scintillator light output for various secondary charged ions are needed [32]. The techniques [5] used in SCINFUL to compute secondary-particle production could be expanded and extended for E_n to ~140 MeV; however, careful experimental response measurements should certainly be made to ensure that the resulting calculations are reasonable.

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